Optical Resolution of the Lamb Shift in Atomic Hydrogen by Laser Saturation Spectroscopy

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This article describes a successful attempt to resolve the single fine structure components of the hydrogen line H_α. The Lamb shift has been observed directly in the optical absorption spectrum.

In this article we report a successful attempt to eliminate Doppler broadening of the red Balmer line H_α of atomic hydrogen at 6563 Å by high resolution saturation spectroscopy[1] using a repetitively pulsed narrow-band tunable dye laser[2]. We have been able to resolve for the first time single fine structure components of H_α and to observe the Lamb shift[3,4] directly in the optical absorption spectrum. The hydrogen was excited in a simple Wood discharge tube[5] and no cooling was necessary. It was possible to observe the saturated absorption in the afterglow and thus to minimize Stark broadening and shifts due to electric fields in the plasma.

Fine Structure

Hydrogen has always attracted the attention of spectroscopists, because its simplicity permits detailed and accurate comparison with theoretical models. Unfortunately Doppler broadening is particularly large for light hydrogen—nearly 6,000 MHz at room temperature—and masks the finer details of the important fine structure of the spectral lines. The result is that accurate knowledge of this structure is at present almost entirely based on radiofrequency spectroscopy and level crossing techniques[4,6], but nevertheless there have been countless efforts to study with increasing resolution the lines of atomic hydrogen, and in particular the Balmer lines in the visible[4]. An accurate measurement of the wavelength is the basis for the precision determination of the Rydberg constants, one of the cornerstones in the evaluation of the fundamental constants[7].

All the reported improvements in resolution have been based on a reduction of the Doppler width, but even if the hydrogen discharge is cooled by liquid H_2 or liquid He, the red Balmer line appears only as a simple doublet, each component being a blend of different fine structure lines. The heavier isotopes D and T, with their reduced Doppler width, permit somewhat better resolution, which has been utilized in a recent experiment[8] by Kibble *et al.* It has not been possible, however, to isolate single fine structure components, so that deconvolution procedures remain necessary in the evolution of wavelength. Experiments to reduce the Doppler width of the Balmer lines by using a collimated atomic beam[9] have been only moderately successful because of poor signal-to-noise ratio. The recently developed narrow-band tunable dye lasers together with the new powerful non-linear spectroscopic techniques have now, however, put us in a much stronger position to meet this challenge, and the preliminary results reported in this article seem to justify our optimism.

The technique used in our experiment is essentially the same as in a recent study of the sodium D lines[1]. A prerequisite is a narrow-band laser, whose frequency can be tuned continuously across the spectral region of interest; its output is split into a weak probe beam and a stronger, periodically blocked saturating beam, which are sent in nearly opposite directions through the absorbing gas medium. The saturating beam can bleach a path for the probe and decrease its absorption, if the laser is tuned close to an atomic resonance frequency so that both lightwaves interact with the same atoms, that is, those with essentially vanishing axial velocity. The saturation spectrum is obtained by recording the probe intensity changes due to the inhomogeneous saturation as a function of frequency.

We used a continuously tunable dye laser[2], pumped by a pulsed nitrogen laser with a repetition rate of 80 p.p.s. The active medium is a solution of cresyl violet (2 g $l.^{-1}$) and rhodamine 6 G (1.2 g $l.^{-1}$) in ethanol[10]. The pulse length is 3 ns. An external piezoelectrically tunable confocal pass band filter reduces the band-width from 0.01 cm^{-1} to 0.001 cm^{-1} or 30 MHz and stretches the pulse length to 8 ns FWHM.

In order to observe any absorption on the hydrogen alpha line, the lower states with $n=2$ (see Fig. 1) must be populated; this is most easily done in a gas discharge. Previous estimates indicated that it should be possible to obtain a sufficient number in a rather gentle discharge, which keeps uncontrolled environmental effects small[3]. In our experiment we used a discharge tube closely resembling an old design originally suggested by Wood[5]. A continuous stream of electrolytically generated H_2 is pumped through a folded Pyrex tube which is 1 m long and of inner diameter 8 mm. The pressure is monitored by a thermogauge and can be varied between 0.1 and 1 torr. A d.c. discharge with a typical current of 10 to 20 mA is maintained between neon sign electrodes at the ends—the voltage across the electrodes is about 1.3 kV at a typical pressure of 0.2 torr. Small traces of water vapour effectively prevent the catalytic recombination of hydrogen atoms at the glass walls and permit a high percentage of dissociation. A central section of the discharge (15 cm long) is probed axially through flat end windows cemented under a slight tilt angle to short tube extensions. Under these conditions a total absorption in excess of 50% is readily obtained, corresponding to an estimated population of 10^{10} cm^{-3} of the state $n=2$. For absorption studies in the afterglow, a vacuum pentode 807 is used as a fast electronic switch in parallel to the discharge. It can interrupt the discharge within 0.1 μs by short circuiting the power supply.

As in the previous experiment[1], the intensity of the probe is compared with that of a second probe, which does not cross the bleached region. Both are about 1 mm in diameter and 5 mm apart. A spatial filter reduces the amount of spontaneous emission falling onto the photodetectors. The output of the phase sensitive amplifier is normalized to constant average probe intensity by means of an electronic analogue divider, and plotted as a function of the tuning voltage.

Saturation Spectrum

Fig. 2 shows a saturation spectrum of the hydrogen alpha line obtained at a H_2 pressure of 0.23 torr in the afterglow of a d.c. discharge with a current of 20 mA. The elapsed time between termination of the discharge and optical observation

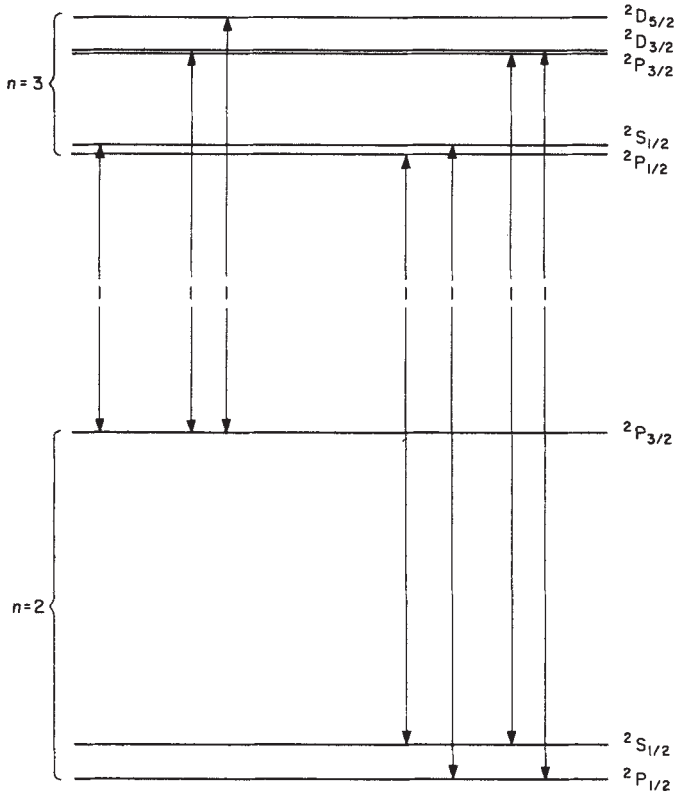

Fig. 1 Energy level of the hydrogen Balmer line H_α with theoretical fine structure[11].

is 1 μs. The total scanning time was 15 min at 3 s integration time constant. The peak powers of probe and saturating beam were reduced to about 5 and 100 mW respectively to keep power broadening small. The laser power was higher at the right half of the spectrum to provide a better signal-to-noise ratio for the weaker components. The theoretical fine structure, as expected from the energy levels of Fig. 1, is plotted for comparison in Fig. 2*b*. The line heights give the relative oscillator strengths[4]. The four strongest components are clearly resolved in the saturation spectrum. Higher laser intensities or higher discharge currents are required to observe the weaker components at the outer left. The positions of the observed resonances are in reasonable agreement with the expected ones, although a precision calibration of the frequency scale, by means of a long, thermally stabilized optical cavity, for example, would be required for an accurate quantitative comparison. The fact that the lines originating in the metastable S state are not more intense seems to indicate that the S and P levels are approximately equally populated. Apparently the short natural lifetime of 1.6 ns for the 2P state is effectively lengthened by resonance trapping of the emitted ultraviolet Lyman alpha radiation. Time dependent absorption measurements in the afterglow show that the life of the metastable 2S state, on the other hand, is reduced to about 1 μs, presumably by collisions and residual fields. The splitting of the clearly resolved doublet at the extreme right is essentially a consequence of the Lamb shift (that is, the fact that the $2^2S_{1/2}$ state is energetically about 1,058 MHz above the $2^2P_{1/2}$ state, by contrast with the Dirac theory).

In saturation experiments of the kind we have performed, additional cross-over signals can occur half-way between two lines that share a common upper or lower level[1,12]. These positions are marked by arrows in Fig. 2*c* and at least one strong resolved component agrees with the position of such a cross-over due to the common lower $2^2S_{1/2}$ state. If the saturation spectrum is recorded while the discharge is on, an additional cross-over line tends to show up in between the two components on the extreme right, even though these transitions have no level in common. It seems that the electric field of about 10 V cm^{-1} in the positive column of the discharge effectively mixes the closely spaced $3^2D_{3/2}$ and $3^2P_{3/2}$ states and thus leads to a breakdown of the ordinary selection rules.

The narrowest observed line-width is about 250 to 300 MHz FWHM for all resolved components. This width is obtained at low bleaching intensity and seems quite insensitive to changes in gas pressure or discharge current. This represents a resolution of 5 parts in 10^7 and is about an order of magnitude narrower than the best previous value, obtained by conventional emission spectroscopy from a cooled deuterium discharge[8]. For lines starting in the 2S state, unresolved hyperfine splitting of 177 MHz (ref. 13) can account for most of the observed width. The hyperfine splitting of the $2^2P_{1/2}$ and $2^2P_{3/2}$ state is smaller (60 and 24 MHz respectively), but the short life of these levels contributes about 100 MHz to the natural line-width. Additional contributions to the experimental width arise from the finite laser band-width and from residual Doppler broadening due to a finite crossing angle (about 0.005 radians) between the two light beams. Obviously the experimental resolution is sufficiently close to the theoretical limit to rule out large contributions from pressure broadening or Stark broadening due to the electrons and ions of the plasma[14].

Close inspection of Fig. 2*a* reveals a low, broad pedestal at the bottom of the two outer strong components. This pedestal is much more pronounced at higher laser intensities. Such a pedestal is expected in the presence of some strong cross-relaxation mechanism that redistributes the atomic velocities[15]. Resonance trapping clearly provides such a mechanism, and may cause difficulties if observation under steady state condition is attempted.

G. W. Series concludes his recent review article[7] on the determination of the Rydberg constant with the remark: "In a more speculative vein one might contemplate the possibility of using hydrogen atoms in the metastable $2^2S_{1/2}$ state as absorbers in a tunable laser locked to some component of the Balmer alpha line. Such a device would allow one to take

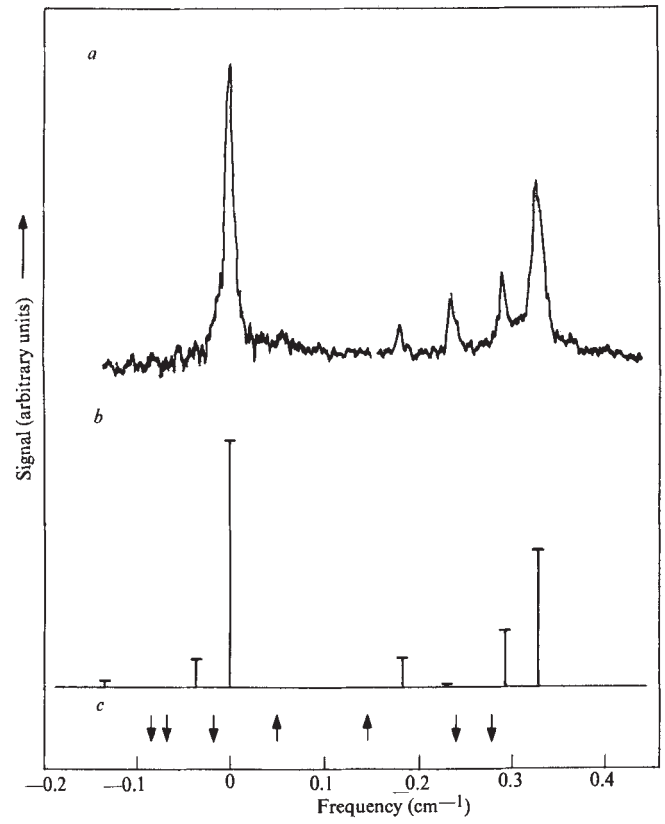

Fig. 2 *a*, Saturation spectrum of H_α; *b*, theoretical fine structure with relative oscillator strength; *c*, position of possible cross-over signals.

advantage of the precision in wavelength determination which is offered by lasers of this type". Our experiment demonstrates that such a proposal is no longer a speculation, but is within the realm of the immediately possible. If the centre of the line can be located within 4% of the width, the Rydberg constant could be determined to an accuracy of one part in 2×10^8, that is, to the accuracy of the present standard of length.

It will also be possible to compare accurately the wavelengths of different Balmer lines and to verify the existence of small, theoretically predicted level shifts[16]. Moreover, the isotope shifts between H, D and T can now be determined with unexcelled accuracy.

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Simplified Photon Counting System for Measurement of Low Radiation Levels

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This article shows that the availability of photomultipliers with gains of about 2×10^6 to 10^8 has obviated the need for an amplifier and that a system having no amplifier and a cooled photomultiplier does not require a pulse height discriminator for efficient operation.

THE continuing development of atomic spectral analysis, particularly in the field of non-flame atom reservoirs, has resulted in a need for more sophisticated techniques of light measurement. The photomultiplier is still the principal detector, but the methods of handling the resultant signal have diversified considerably recently, for example, d.c. amplification/integration, coherent detection, time averaging. Photon counting is by definition the most sensitive method for the detection of radiation in the ultraviolet-visible region and is at the same time the simplest and most obvious because it involves the direct recording of the inherently digital output of the photomultiplier. Some of the more important properties of photon counting are as follows. (1) All photon counts include signal, background and noise photons; (2) the light signal is integrated over the count period which allows the detection of extremely low light levels, 10 photo-electrons s^{-1}, for example; the equivalent d.c. current from a photomultiplier with a gain of 10^7 would be about 1.6×10^{-11} A with a standard deviation of about 4×10^{-12} A; (3) dark pulses from the photomultiplier can be removed by cooling and/or pulse height discrimination; (4) reading errors associated with analogue techniques are eliminated; (5) the integral nature of the measurement is useful when transient radiation signals of unknown profile[1] are to be measured, or when the signal has poor short term stability.

The basic principles of photon counting together with several practical systems and their application to absorption spectrophotometry and atomic spectrometry have been described[2-6]. Each practical system has comprised five basic components, namely the photomultiplier, the photomultiplier anode load, a fast amplifier, a pulse height discriminator and a digital counter (frequency meter).

Photon Counting System

The chief function of the discriminator as used in our previous studies was to remove amplifier noise and not dark current pulses from the photomultiplier. Hence the pulse heights derived from an EMI 9601B photomultiplier (gain about 2×10^6) were measured to determine the usefulness of the amplifier. Because no pulse height analyser was available, the measuring circuit used was a Tektronix type 547 oscilloscope fitted with a dual trace plug-in (type 1A1) and probe (10 mΩ, 7 pF, ×10, No. P6008). The probe was attached directly to the end of the output cable from the photomultiplier (length 6 inch). Obviously this cable must be kept as short as possible because its capacity is combined with the total stray capacity of the photomultiplier anode circuit C_s which, together with the anode load resistance R_1, form the anode time constant C_sR_1 (ref. 5). The magnitude of C_sR_1, as shown later, determines the maximum pulse repetition frequency that the photon counter can handle. Photo-electric pulses generated by photomultipliers have been shown to have an approximately Poisson distribution of pulse heights which made visual estimates of pulse size extremely difficult. Under these conditions three estimates of pulse size were made to ensure greater consistency. The results obtained are shown in Table 1.

The digital counter (frequency meter) used (Venner Electronic, model TSA 6636/3) had a 10 mV counting threshold and therefore the EMI 6256S photomultiplier (gain of 4×10^7) was considered suitable. Although the use of the probe