Doppler-Free Two-Photon Spectroscopy of Hydrogen $1S$-$2S$*

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We have observed the $1S$-$2S$ transition in atomic hydrogen and deuterium by Doppler-free two-photon spectroscopy, using a frequency-doubled pulsed dye laser at 2430 Å. Simultaneous recording of the absorption spectrum of the Balmer-β line at 4860 Å, using the fundamental dye-laser output, allowed us to precisely compare the energy intervals $1S$-$2S$ and $2S,P$-$4S,P,D$ and to determine the Lamb shift of the $1S$ ground state to be 8.3 ± 0.3 GHz (D) and 8.6 ± 0.8 GHz (H).

We have observed transitions from the $1S$ ground state of atomic hydrogen and deuterium to the metastable $2S$ state, using Doppler-free two-photon spectroscopy.[1-3] The atoms are excited by absorption of two photons of wavelength 2430 Å, provided by a frequency-doubled pulsed dye laser, and the excitation is monitored by observing the subsequent collision-induced $2P$-$1S$ fluorescence at the L_α wavelength 1215 Å. Linewidths smaller than 2% of the Doppler width were achieved with two counter-propagating light beams, whose Doppler shifts cancel. The fundamental dye-laser wavelength at resonance 4860 Å coincides with the visible Balmer-β line, and simultaneous recording of the absorption profile of this line permits a precise comparison of the energy intervals $1S$-$2S$ and $2S,P$-$4S,P,D$. From our first preliminary measurements we have determined the Lamb shift of the $1S$ ground state to be 8.3 ± 0.3 GHz (D) and 8.6 ± 0.8 GHz (H), in good agreement with theory. The only previous measurement of the Lamb shift of the $1S$ state of deuterium, 7.9 ± 1.1 GHz, has been reported by Herzberg,[4] who used a difficult absolute-wavelength measurement of the L_α line. The hydrogen-$1S$ Lamb shift has never been measured before.

Numerous authors[2,3,5] have pointed out that it would be very desirable to observe the $1S$-$2S$ transition in hydrogen by Doppler-free two-photon spectroscopy. The $\frac{1}{7}$-sec lifetime of the $2S$ state promises ultimately an extremely narrow resonance width. The resolution obtained in the present experiment is already better than that achieved in our recent study of the Balmer-α line by saturation spectroscopy,[6] and the implications for a future even more precise measurement of the Rydberg constant are obvious.

We utilized a dye-laser system, consisting of a pressure-tuned dye-laser oscillator with optional confocal-filter interferometer[7] and two subsequent dye-laser amplifier stages, pumped by the same 1-MW nitrogen laser (Molectron UV 1000) at 15 pulses/sec. This laser generates 10-nsec-long pulses of 30–50-kW peak power at 4860 Å with a bandwidth of about 120 MHz (1–2 GHz without confocal filter). A 1-cm-long crystal of lithium formate monohydrate (Lasermetrics) generates the second harmonic with a peak power of about 600 W. A detailed description of this laser system will be published elsewhere.

The ground-state hydrogen atoms are produced by dissociation of H_2 or D_2 gas in a Wood-type discharge tube (1 m long, 8 mm diam, typically 0.1–0.5 Torr, 15 mA). The atoms are carried by gas flow and diffusion through a folded transfer tube about 25 cm in length into the Pyrex observation chamber (Fig. 1). This chamber has two side arms with quartz Brewster windows to transmit the uv laser light and a MgF_2 (originally LiF) window for the observation of the emitted L_α photons. A thin coating of syrupy phosphoric acid is applied to all Pyrex walls to reduce the catalytic recombination of the atoms.

The uv laser light is focused into the chamber

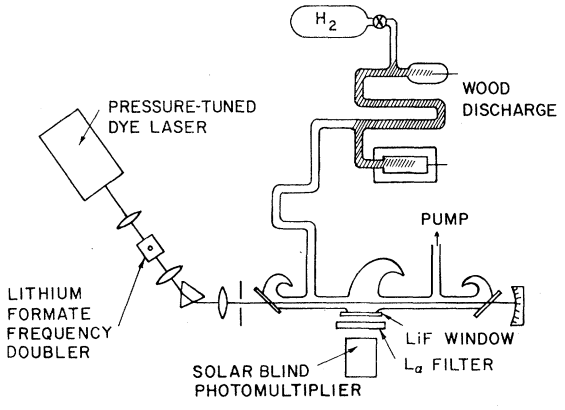

FIG. 1. Experimental setup.

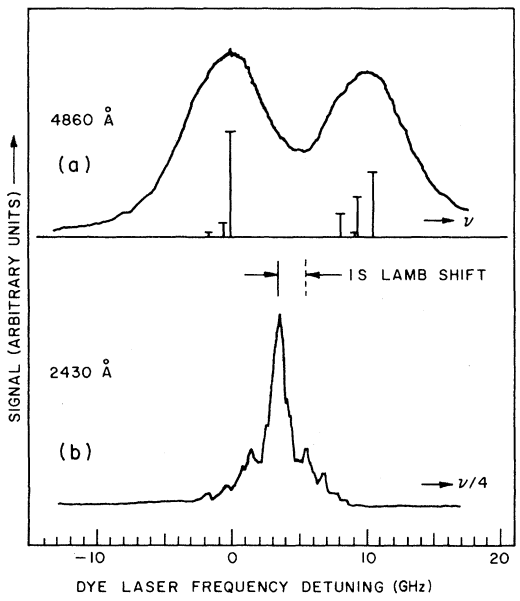

FIG. 2. (a) Absorption profile of the deuterium Balmer-β line with theoretical fine structure; (b) simultaneously recorded two-photon resonance of deuterium $1S$-$2S$.

to a spot size of about 0.5 mm diam. The transmitted beam is refocused into the cell by a spherical mirror to provide a standing-wave field. The separation between illuminated region and MgF_2 window is kept small (1–2 mm) to reduce the loss of L_α photons due to resonance trapping.

The transmitted L_α photons are detected by a solar-blind photomultiplier (EMR 541 J). An interference filter with 6% transmission at 1215 Å reduces the off-resonance background signal to less than one registered photon per several hundred laser pulses. The multiplier output is processed by a gated integrator (Molectron LSDS) with an effective gate opening time of 200 μsec and is electronically divided by a signal proportional to the square of the uv laser intensity.

Figure 2(b) shows a two-photon spectrum of deuterium $1S$-$2S$ recorded with moderate resolution (no confocal-filter interferometer). The low Doppler-broadened pedestal is caused by two-photon excitation by each of the linearly polarized uv beams individually and could be eliminated by the use of circularly polarized light.[3] The signal at resonance corresponds to about 10–20 registered L_α photons per pulse, and remains within the same order of magnitude when the H_2 or D_2 gas is diluted by He up to a ratio of 1000:1. The expected decrease in the number of excited metastable atoms is apparently largely compensated over a wide range by a concomitant reduction of the loss of L_α photons due to resonance trapping and quenching. Despite the lack of a near-resonant intermediate state, the two-photon fluorescence is comfortably strong, and it should easily be possible to observe the signal at a considerably lower total gas pressure (10^{-4} Torr), where pressure broadening and shifts would become unimportant if the $2S$-$2P$ transitions were induced by an applied rf field.

Figure 2(a) shows the absorption spectrum of the deuterium Balmer-β line which was simultaneously recorded by sending a small fraction of the blue dye-laser light through a 15-cm-long center section of the positive column of a Wood-discharge tube (0.2 Torr D_2, 25 mA). The positions of the indicated theoretical fine-structure components were located by a computer fit of the line profile. We have measured the separation of the $1S$-$2S$ resonance from the strongest component ($2P_{3/2}$-$4D_{5/2}$) in the Balmer-β spectrum to be 3.38 ± 0.08 GHz for deuterium and 3.3 ± 0.2 GHz for hydrogen (in terms of the blue-light frequency). The corresponding theoretical separations[8] are 3.420 and 3.422 GHz, respectively. These separations would be larger if the $1S$ state were not raised above its Dirac value by the Lamb shift (theoretically 8.172 GHz for D and 8.149 GHz for H) and our measurement can be interpreted as a determination of the ground-state Lamb shift. A considerable improvement in accuracy can be expected when a high-resolution saturation spectrum[6] of the Balmer-β line is used for the comparison.

A two-photon spectrum of hydrogen $1S$-$2S$ with the laser operating in its high-resolution mode is shown in Fig. 3 (scan time about 2 min). The linewidth is limited by the laser bandwidth of about 120 MHz (in the blue). The spectrum reveals two hyperfine components, separated by the difference of the hyperfine splittings of lower and upper states, as expected from the selection rule $\Delta F = 0$.[2]

It is not difficult to compare the observed signal strength with theoretical estimates, using Eq. (7) of Ref. 2, derived for steady-state conditions. In the present experiment the atoms are excited by light pulses whose time duration τ is short compared to the inverse linewidth Γ_e of the two-photon transition, and which have a near-Fourier-transform-limited bandwidth $\Delta\omega \approx \tau^{-1}$. One can show with the help of time-dependent perturbation theory[9] that Eq. (7) of Ref. 2 in this case still

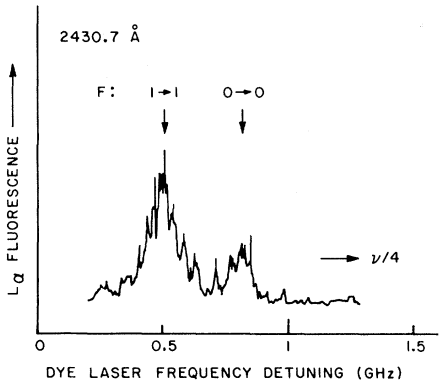

FIG. 3. High-resolution two-photon spectrum of hydrogen 1S-2S with resolved hyperfine splitting.

correctly predicts the effective two-photon transition rate if one replaces the linewidth Γ_e by the laser bandwidth $\Delta\omega$. We have numerically evaluated this expression in a calculation similar to that of Ref. 5, but with inclusion of the continuum in the summation over intermediate states, and obtain a two-photon transition rate per atom of $\Gamma \approx 7\times10^{-4} I^2/\Delta\omega$, where the light intensity I is measured in W/cm^2, the laser bandwidth $\Delta\omega$ in MHz, and Γ in sec^{-1}.

For a comparison with the experiment we consider a H_2 partial pressure of 2×10^{-4} Torr and 10% dissociation, i.e., a density of 1.4×10^{13} 1S atoms/cm^3, so that the loss by resonance trapping is not important. Our estimate then predicts about 3×10^5 excited metastable atoms per pulse over a 1-cm path length. With a detection solid angle of $0.02\times4\pi$ sr, a filter transmission of 6%, and a multiplier quantum efficiency of 10%, we expect about thirty registered photons per laser pulse, in reasonable agreement with the present observations.

We have also calculated the shift of the 1S-2S two-photon resonance frequency caused by the intense uv radiation (ac Stark effect). By numerically evaluating Eq. (1) of Liao and Bjorkholm,[10] we estimate an intensity shift of about 5.5 Hz/(W/cm^2) or less than 2 MHz under the present operating conditions. The light intensity and hence the shift can, in principle, be reduced without loss of resonance signal, by decreasing the laser bandwidth and increasing the interaction time with the atoms. A hundredfold improvement in resolution should be obtainable with the present pulsed dye-laser system, if the hydrogen cell is placed inside a narrow-band confocal-filter interferometer.

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