Continuous-Wave Two-Photon Spectroscopy of the 1S-2S Transition in Hydrogen

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We have observed the $1S$-$2S$ transition in atomic hydrogen gas with a resolution of 5 parts in 10^9 by Doppler-free two-photon spectroscopy, using multimilliwatt cw radiation near 243 nm generated by sum-frequency mixing. The experiment opens the way to substantial increases in the accuracy of the Rydberg constant, the electron/proton mass ratio, and the Lamb shift of the hydrogen $1S$ ground state.

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We have, for the first time, observed the $1S$-$2S$ transition in atomic hydrogen gas by continuous-wave Doppler-free two-photon spectroscopy. To do this, we employed cavity-enhanced multimilliwatt radiation near 243 nm generated by sum-frequency mixing. The resolution of 5 parts in 10^9 surpasses earlier pulsed experiments[1] by more than an order of magnitude, and dramatic further improvements are expected from experiments with a cold hydrogen beam. Measurements of the absolute frequency and the hydrogen-deuterium isotope shift and comparisons with other sharp transitions in hydrogen promise accurate new values of fundamental constants and stringent tests of quantum electrodynamic computations.

Ever since it was discovered that first-order Doppler broadening can be eliminated by two-photon excitation with counterpropagating laser beams,[2] hydrogen $1S$-$2S$ has been recognized as one of the most intriguing transitions to be studied by high-resolution laser spectroscopy. Unlike the corresponding transition in postronium,[3] its resolution is not limited by annihilation, and the $\frac{1}{8}$-sec lifetime of the metastable $2S$ state promises an ultimate resolution of better than 1 part in 10^{15}.

The linewidth in past experiments has been entirely instrument limited, since intense tunable radiation at the required ultraviolet wavelength near 243 nm was only available from pulsed laser sources. Wieman[1] reached a resolution of 1.3 parts in 10^7 with the help of a cw dye laser oscillator at 486 nm, followed by a pulsed dye amplifier and lithium formate frequency doubler. A comparison with the $n=2$-4 Balmer-β transition yielded the $1S$ Lamb shift with an accuracy of 0.4% or 30 MHz, limited by frequency chirping in the pulsed dye amplifiers. Very recently, Hildum[4] has recorded new pulsed $1S$-$2S$ spectra in a hydrogen atomic beam, detecting the signal via photoionization of the $2S$ atoms. A passive filter interferometer in between the pulsed dye amplifier and a urea frequency doubler provides a resolution of 5 parts in 10^8, permitting a more accurate comparison with Balmer-β and promising an improved value of the Rydberg constant[5] from a comparison of the fundamental dye laser wavelength with a nearby $^{130}Te_2$ reference line whose absolute wavelength has recently been measured[6] within 4 parts in 10^{10}.

It has long been obvious that only a cw source of intense, highly monochromatic radiation at 243 nm can do justice to the extremely narrow natural linewidth of the $1S$-$2S$ transition. Unfortunately, there are no tunable lasers which operate directly at this wavelength. Attempts to double the frequency of a cw dye laser with angle-tuned crystals of lithium formate or urea inside the cavity have been plagued by crystal damage,[7] and no nonlinear optical crystal is known which would permit efficient 90° phase-matched second-harmonic generation at 243 nm. Phase-matched sum-frequency mixing of two different primary wavelengths is possible,[8,9] however, and uv powers as high as 4 mW have recently been generated[8] by summing the frequency of a 364-nm argon laser and a 732-nm dye laser in an ammonium dihydrogen phosphate (ADP) crystal cooled to $-25\,^{\circ}C$. The intensities of both primary beams at the crystal were enhanced with servo-locked passive ring cavities. Unfortunately, the cold ADP surfaces are susceptibile to damage by moisture, and thermal lens effects created in the crystal by the red light severely limit the enhancement and stability of the second cavity.

A very simplified scheme of the present experiment is shown in Fig. 1. The cw source is similar to that described in Ref. 8, but the reliability and ease of use are much improved by sum-frequency mixing in potassium dihydrogen phosphate (KDP) rather than ADP. Efficient 90° phase matching is possible with the crystal heated to 62 °C, when summing the ouput of a 351-nm argon laser and a 790-nm dye laser. We employ an argon laser (Coherent I-20) with a prism wavelength selector and intracavity etalon,[8] and an LD-700 cw ring dye laser (Coherent 699-21) pumped by 8 W from a red krypton-ion laser. Each primary laser provides about 500 mW of single-frequency radiation with a linewidth of 1 MHz, servo locked to a passive reference cavity. The KDP crystal (45° z cut, $8\times8\times25$ mm^3), with faces at Brewster's angle, is mounted inside a small windowless housing, temperature stabilized to within 0.01 °C. The dye-laser intensi-

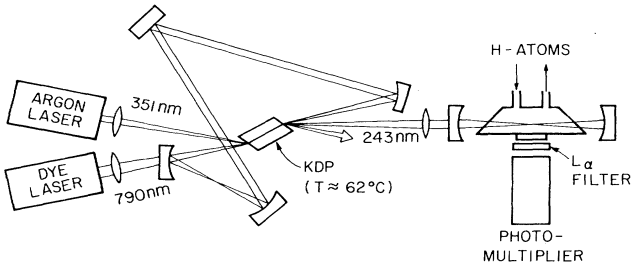

FIG. 1. Simplified scheme of the apparatus for cw two-photon spectroscopy of hydrogen $1S$-$2S$.

ty at the crystal is enhanced with a passive ring cavity, servo locked by monitoring the polarization of the reflected light.[8] The excellent surface polish obtainable with KDP permits a 30–50-fold intensity enhancement, providing a circulating red power of 15–25 W. When the argon laser beam has a waist size of 30 μm and the focusing of the red cavity is adjusted for maximum power, the sum-frequency output reaches 1–2 mW even without cavity enhancement of the argon-laser light. Stable operation without degradation in power is now routinely achieved over periods of many hours. Mode-matching lenses guide the 243-nm radiation into a servo-locked standing-wave cavity (length 23.7 cm, mirror radius 25 cm, reflectivity 96.5%, waist size 100 μm), which enhances the ultraviolet intensity about eightfold in the final interaction region.

The observation cell inside the cavity is equipped with crystalline quartz Brewster windows for the 243-nm radiation. The hydrogen atoms are produced by dissociation of H_2 in a flowing-gas dc discharge tube,[1] and reach the cell by traveling through a 24-in.-long Teflon hose. The signal is detected by monitoring collision-induced Lyman-α emission through a LiF side window and vacuum-uv interference filter with a solar-blind photomultiplier (EMR 541J) followed by photon-counting electronics. With a solid angle of 1%, a filter transmission of 15%, and a cathode efficiency of 10%, the detection efficiency is about 1.5×10^{-4} before accounting for losses caused by resonance trapping and radiationless quenching.

Figure 2 shows a two-photon spectrum of hydrogen $1S$-$2S$ with its two hyperfine components, recorded in this way at a gas pressure of approximately 0.2 Torr and a 243-nm power of about 10 mW traveling in each direction inside the cavity. The argon-laser frequency was kept fixed, while the dye laser was scanned at a rate of about 1 MHz/sec. The output of the photon counter (integration time 1 sec) was simply sent to a chart recorder or digital data logger, without employing any modulation techniques, normalization, or further signal smoothing. The signal at the large $F=1$ resonance reaches 1000 counts/sec. The residual background is almost entirely due to 243-nm stray light, and a large improvement in signal-to-noise ratio can be

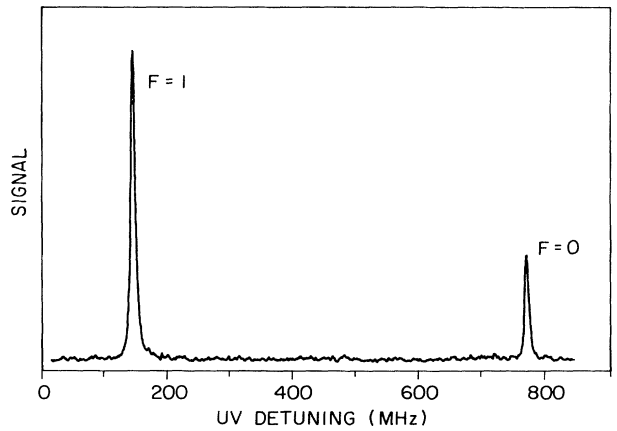

FIG. 2. Doppler-free two-photon spectrum of hydrogen $1S$-$2S$. The atoms are excited by 10 mW of cw radiation at 243 nm, and the signal is monitored by counting collision-induced vuv Lyman-α photons.

expected from the installation of simple light baffles.

The resonance width of about 8 MHz (full width at half maximum at 243 nm) in Fig. 1 is somewhat broadened by collisions (≈ 2 MHz) and narrower lines have been observed at lower gas pressures. The laser linewidth is estimated to contribute another 2 MHz to the recorded width. The largest source of line broadening is the short transit time of the hydrogen atoms moving through the ultraviolet beam waist.

All these sources of line broadening can be greatly reduced by straightforward means. The instrumental linewidth can be reduced into the subkilohertz range with the help of internal and external laser frequency stabilizers with a faster servo response.[10] Collision broadening can be avoided with an atomic beam; and transit-time broadening can be reduced by sending the atoms along the light waves, or by Ramsey-fringe spectroscopy with two spatially separate interaction regions.[11] With a hydrogen beam at room temperature and an interaction length of 1 cm, transit broadening can thus be kept below 60 kHz, comparable to the second-order Doppler shifts due to the time dilatation of special relativity (70 kHz). Much improvement is gained if the hydrogen beam is cooled to about 4 K by mounting the escape nozzle on a liquid-helium cryostat.[12] In our example, transit broadening will then be reduced to 7 kHz, and second-order Doppler shifts remain smaller than 1 kHz. At the same time, the longer interaction time due to cooling increases the excitation probability per atom almost eightyfold (from about 10^{-9} to 8×10^{-8} for a light field of 10 mW, focused to a waist of 0.1 mm).

For experiments in the more distant future, we are exploring ways to slow the hydrogen atoms far below liquid helium temperature. One promising approach may be laser-radiation–pressure cooling.[13] A hydro-

gen atom at 4 K can be stopped by resonant scattering of only 100 Lyman-α photons. As an alternative, the large magnetic moment of the atoms could be used for velocity reduction and selection in inhomogeneous magnetic fields. Once the kinetic energy has been reduced to the millikelvin range, gravity can further slow the atoms in a "hydrogen fountain" of modest dimensions.[14] Ramsey-fringe signals could be recorded in such a fountain with a single laser field by exciting the atoms near their turning point on their way up and again on their way down. Unlike in the standard Ramsey setup, the fastest atoms contribute the narrowest fringes in such a fountain experiment, and when averaging over a Maxwellian velocity distribution we expect a near-Lorentzian signal whose width equals the natural linewidth. A resonance width of 1 Hz should thus ultimately be achievable in this way.

But even the much more limited resolution of the present experiment is opening interesting opportunities for new precision measurements. The absolute frequency of the $1S$-$2s$ transition can be compared with that of the recently measured $^{130}Te_2$ reference line[6] by locking a 486-nm-cw dye laser to this line and observing an rf beat signal between its second harmonic and the output of the sum-frequency generator. A considerably more elaborate frequency chain could be designed to compare the $1S$-$2S$ frequency directly with the microwave cesium frequency standard. Such a measurement can yield a much improved Rydberg value and thus improve the link between the time standard and the system of fundamental constants. However, it cannot directly serve as a test of fundamental physics laws, since the cesium hyperfine frequency is not well understood theoretically.

It appears more interesting to compare the $1S$-$2S$ transition with another optical frequency that can also be calculated from first principles, such as another transition in the hydrogen atom itself. An obvious candidate is the Balmer-β line at 486 nm, used in previous measurements of the $1S$ Lamb shift. However, the large natural linewidth (≈ 14 MHz) of even the narrowest fine-structure components limits the potential accuracy. Considerably narrower linewidths (≈ 2 MHz) have recently been observed by Doppler-free two-photon excitation of $8S$ and $8D$ Rydberg states in a beam of metastable $2S$ atoms.[15] Two-photon spectroscopy of higher $2S$-nS transitions allows even better resolution, if perturbing external fields can be eliminated. Rydberg levels close to the ionization limit require a laser wavelength near 729 nm, slightly more than 3 times larger than the 243 nm of the $1S$-$2S$ resonance, and it should be feasible to compare the two laser frequencies with extreme precision by detecting a microwave beat signal between the ultraviolet light and the third harmonic of the red laser.

These new opportunities make it worthwhile to examine in some detail what can be learned from a comparison of experiment and theory. Although quantum electrodynamics is believed to provide a very good theory for the hydrogen atom, the accuracy of calculated energy levels[16] is limited by the uncertainty of fundamental constants, by unknown nuclear size and structure effects, and by computational approximations. However, important cancellations of these uncertainties can occur if different transition frequencies are compared. In order to facilitate the analysis of theoretical uncertainties, we have adapted Erickson's model[16] to a microcomputer spreadsheet program (Lotus 1-2-3), which permits interactive changes of constants and parameters, computing term energies and transition frequencies to fifteen-digit numerical accuracy within a few seconds.

The dominant contribution to the uncertainty of the $1S$-$2S$ interval (2.5 MHz) is the uncertainty of the Rydberg constant[5] (10^{-9}), and an absolute wavelength or frequency measurement can improve this important constant about tenfold, until we approach the uncertainties due to the electron mass[16] (5×10^{-8}: 70 kHz), the charge radius of the proton[16] (10^{-1}: 130 kHz), and approximations in the computation of QED corrections (60 kHz). The fine-structure constant (10^{-7}) contributes an uncertainty of only 4 kHz.

Measurements of the $1S$-$2S$ hydrogen-deuterium isotope shift have previously been suggested as a means to measure the electron/proton mass ratio.[1] However, after recent improved measurements of this ratio,[17] the uncertainty of the isotope shift is no longer dominated by the electron mass (35 kHz), but instead by nuclear size effects (180 kHz).

A comparison of $1S$-$2S$ with a Rydberg transition $2S$-nS can provide the measurable small difference frequency $d\nu = \nu(1S\text{-}2S) - 3\nu(2S\text{-}nS)$ which depends critically on the Lamb shifts of the participating levels and provides a stringent test of QED. For $n \approx 100$, its theoretical uncertainty is dominated by the nuclear size effect (≈ 70 kHz) and by approximations in the computation of electron structure corrections and uncalculated higher-order QED corrections[16] (65 kHz). The contributions of the Rydberg constant (1 kHz), the electron mass (0.05 kHz), and the fine-structure constant (4 kHz) are negligible, by comparison. The QED computations can be improved, and if the theory is correct, a precision measurement of $d\nu$ can provide accurate new values for the charge radii of the proton and deuteron.

Finally, we would like to point out that a very interesting composite frequency is obtained by subtracting the correction $d\nu\times 7/(4+24/n^3)$ from the optical frequency $\nu(1S\text{-}2S)$. The multiplication factor is chosen so as to eliminate all energy shifts which scale with the inverse cube of the principal quantum

number. The new frequency is thus independent of the nuclear size, and it no longer depends on uncalculated higher-order QED effects. Measurements of this frequency and its isotope shift can hence provide much improved values of the Rydberg constant and the electron/proton mass ratio.

Very stringent tests of basic physics laws would be possible if better precision values for the same constants could be obtained by independent means. Radio-frequency spectroscopy of transitions between highly excited hydrogen Rydberg levels may provide a promising approach for such measurements.[18] It will also be most interesting to compare hydrogen $1S$-$2S$ with sharp optical transitions in positronium, muonium, pionium, or other hydrogenlike systems.

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