High-Resolution Spectroscopy of the Hydrogen 1S-2S Transition in an Atomic Beam

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We have observed the $1S$-$2S$ transition of atomic hydrogen to a resolution of 5 parts in 10^{11} by means of Doppler-free two-photon spectroscopy in an atomic beam. The significantly asymmetric line shape is strongly affected by the relativistic Doppler effect. The new high resolution can be used for improved measurements of the Rydberg constant, the $1S$ Lamb shift, and the isotope shift of hydrogen and deuterium.

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As the simplest of the stable atoms, hydrogen permits unique confrontations between experiment and quantum electrodynamic theory. The $1S$-$2S$ two-photon transition has long been recognized as one of the most intriguing atomic resonances. Its 1-Hz natural linewidth near 243 nm has become a "holy grail" of high-resolution laser spectroscopy. Recent observations of this transition by continuous-wave Doppler-free two-photon spectroscopy have reached a resolution of about 3 parts in 10^9. Absolute frequency measurements to a few parts in 10^{10} have yielded accurate values for the Rydberg constant and the Lamb shift of the $1S$ ground state.[1,2]

Here we report on the first observation of the $1S$-$2S$ transition in an atomic beam by means of Doppler-free, continuous-wave, two-photon spectroscopy in a longitudinal excitation geometry. Because of the absence of collisions and the long interaction time between the atoms and the laser light, the resonance could be resolved to 5 parts in 10^{11}. This surpasses the resolution of earlier experiments carried out in a gas cell by 2 orders of magnitude so that the $1S$-$2S$ transition is now the best resolved transition of the hydrogen atom.[3-5] The linewidth of 60 kHz at the excitation wavelength near 243 nm is determined to approximately equal parts by the transit time of the atoms passing the laser beam and by the second-order Doppler effect, i.e., by the slowing of the oscillation of moving atoms due to relativistic time dilation. Our experiment gives perhaps the clearest manifestation of this effect to date for slow atoms near room temperature.[6,7] Further improvements in the resolution of the hydrogen transition of 1 to 2 orders of magnitude are expected by cooling the atoms to liquid-helium temperature and by filtering out atoms of high transverse velocity which therefore do not interact with the light over the entire length of the beam. Additional narrowing of the linewidth is possible in a time-resolving measurement in which a time delay between the excitation and the detection inhibits the contribution of the fast atoms to the signal.

Because of its simplicity, the hydrogen atom has played a central role in the development of fundamental laws of physics. Atomic theories have always been extensively applied to it and tested by high-resolution spectroscopy of the predicted energy states.[8,9] The transition from the $1S$ ground state to the metastable $2S$ state with its extremely narrow natural linewidth is one of the most attractive candidates for such tests. It can be driven by Doppler-free two-photon excitation and renders access to a measurement of the $1S$ Lamb shift, which includes a number of fundamental phenomena like the difference of the electron's self-energy in the free and the bound state, the effect of the vacuum polarization on the binding energy, as well as nuclear size effects.

In earlier continuous-wave measurements of the $1S$-$2S$ transition, all carried out in a gas cell, the $1S$ Lamb shift was determined to about 1 part in 10^4.[1,2] These measurements were limited by the instrumental resolution of several MHz, by frequency shifts due to atomic collisions, by the precision of the secondary frequency standard, and by the uncertainty in the knowledge of the Rydberg constant.

To overcome the two latter limitations we are preparing an experiment which does not rely on an absolute frequency measurement but directly compares the fourth part of the $1S$-$2S$ transition frequency to the $2S$-$4S$ frequency. This frequency difference is dominated by the Lamb shifts of the participating levels and a measurement promises to surpass the present precision in the theoretical Lamb shift by a factor of 10.[10] We are also planning a much improved measurement of the isotope shift of the $1S$-$2S$ transition for hydrogen and deuterium. A direct radio-frequency measurement of the 168-GHz frequency difference (at 486 nm) is possible with the help of an electro-optic modulator at 84 GHz.[11] The present resolution of the $1S$-$2S$ transition can be more fully exploited with the help of a laser frequency chain which should permit a comparison of the optical transition frequency with the cesium atomic clock to better than 1 part in 10^{12}. We are also exploring a novel optical frequency divider and synthesizer[12] which should permit precise optical frequency comparison with other sharp transitions in hydrogen, such as $2S$-$8S,8D$.[3] If quantum-electrodynamic theory is correct, such measurements will yield much improved values for the Rydberg constant, the electron-proton mass ratio, and the charge radius of the proton.

Figure 1 shows a simplified scheme of the experimental setup. The coherent exciting light at 243 nm is generated by frequency doubling in β-barium-borate. This crystal, available since 1986, allows direct continuous-wave second-harmonic generation of several mW. In contrast to former experiments, now only one laser is necessary.[2] We use a continuous-wave coumarin-102 dye laser servo locked to an external highly stable Fabry-Perot-type resonator by means of a radio-frequency sideband technique.[13] An additional electro-optic modulator inside the laser cavity serves as a fast servo element to compensate for frequency fluctuations caused by thickness variations of the dye jet. To increase the passive stability, the commercial nozzle was exchanged with one made of highly polished stainless steel with extra sharp edges. In this way a frequency stability of the laser relative to the reference resonator of better than 10 Hz could be achieved for measurement times within 1 ms and 1 s.

To get a good absolute frequency stability, the reference resonator consists of two dielectric mirrors (reflectivity 99.7%, radii 1 m and ∞) cemented on a 46-cm-long Zerodur spacer. It is suspended by a system of Zerodur bars, steel balls, and O rings inside an aluminum cylinder which is finally held by five steel wires inside a vacuum chamber pumped by a ion getter pump to 5×10^{-7} mbar. Thermal and acoustic noise could be suppressed to better than 1 kHz. Only a linear drift with a rate of about 2 kHz/s remains. The laser can be tuned within 300 MHz by means of an acousto-optic modulator (AOM), which shifts the frequency of the laser beam used for the stabilization. For bigger frequency steps the laser has to be locked to different longitudinal modes of the reference resonator. A tellurium spectrometer similar to that in Ref. 2 in combination with an auxiliary piezoelectrically tuned resonator serves to locate the hydrogen frequency within about 5 MHz.

For efficient second-harmonic generation, the fundamental light is enhanced in an external buildup ring resonator, servo locked to the laser by monitoring the polarization of the reflected light.[14] The 200-mW output of the laser is enhanced about 25-fold and about 2 mW uv light is generated in the crystal ($4\times4\times6$ mm^3). The strong astigmatism of the uv light caused by angle-tuned phase matching at 55° is compensated by a tilted curved mirror (radius 10 cm) with an incident angle of about 50°. Mode-matching lenses guide the uv beam into a standing-wave cavity inside the atomic-beam apparatus with both mirrors in vacuum (length 25 cm; waist size 135 μm; input mirror: plane, 4% transmission; output mirror: radius of 50 cm, 0.5% transmission). The resonance of the cavity is stabilized by a second rf sideband servo lock. Routinely, a stable uv power in the cavity of 10 to 15 mW is achieved, which is limited mainly by the antireflection coating of the doubling crystal: If more than 2 mW uv is generated, the coating suffers optical damage which reduces the fundamental intracavity power within minutes. Under optimum conditions we have observed a maximum in the generated uv power of 16 mW. uv-induced optical damage also occurs in the mirror coatings of the uv cavity inside the vacuum chamber if the coating materials contain oxide compounds. Fluoride coatings are therefore mandatory.

The underlying principles for the construction of the atomic-beam apparatus are described in detail by Walraven and Silvera.[15] The hydrogen atoms are produced by dissociation of H_2 in a flowing-gas microwave discharge tube and guided to a nozzle in the vacuum chamber by a 20-cm-long Teflon tube. The nozzle consists of a channel (25 mm length, 1.7 mm diam) in a small aluminum block which can be cooled to liquid-helium temperature. The atoms are injected via another side channel ending on one of the cavity mirrors which simultaneously serves as the rear wall of the nozzle. With this geometry the atoms leave the nozzle mainly in the direction of the laser beam and a long interaction length can be expected. The vacuum chamber (50 cm diam) is pumped by a cryopump (pump rate of 20000 ℓ/s) which allows a pressure of 5×10^{-6} mbar at a hydrogen flowing rate of 5×10^{17}/s.

The excited atoms are detected 15 cm downstream in a "quench zone" where a small electric field (30 V/cm) mixes the $2S$ and $2P$ states, resulting in a fast spontaneous decay of the excited state. A solar-blind photomultiplier (EMR 541I) close to the quench zone detects the Lyman-α photons with an efficiency of $\frac{1}{200}$. The photomultiplier pulses are amplified and counted by a CAMAC module and recorded with a personal computer which also controls the laser frequency. To avoid a strong background due to 243-nm stray light the laser is blocked during the detection for 100 μs by a light chopper. During the excitation period (also 100 μs) the photon counter is disabled, gated by a fast photodiode which monitors the light transmitted by the chopper. This method suppresses the 243-nm background completely. A remaining count rate of about 10/s originates

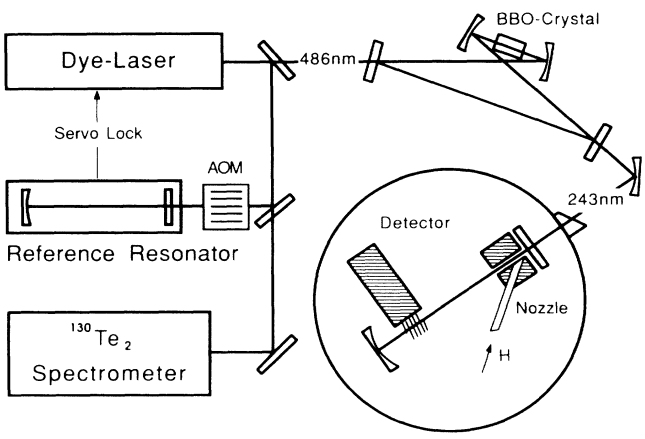

FIG. 1. Schematic overview of the experimental setup.

from the stray light of the discharge.

Figure 2 shows a spectrum of the hydrogen $1S$-$2S$, $F=1$ transition. The laser is scanned in steps of 2.5 kHz with a counting time of 100 ms at each step. The asymmetric form of the measured resonance (solid circles) can be explained as a superposition of exponentially shaped resonances, each for one class of atoms with the same longitudinal velocity. These resonances are transit broadened due to the average transverse velocity but shifted by the relativistic Doppler effect according to the longitudinal velocity of the specific class. The average overall longitudinal velocities lead to a redshifted and asymmetrically broadened line. The solid curve shows the calculated line shape corresponding to an average transverse velocity of 29 m/s and an average longitudinal velocity of 2360 m/s. Supposing a Maxwellian velocity distribution the mean longitudinal velocity is equivalent to a temperature of 200 K which is in reasonable agreement with the measured nozzle temperature of 170 K at which the line was recorded. From the average transverse velocity a mean interaction time of 9 μs can be derived corresponding to a mean interaction length of 2 cm. This yields a 1° angular spread of the detected metastable beam. From the count rate of 3000/s and the detection efficiency of the photomultiplier we estimate an intensity of 6×10^5 metastable atoms per second. The dashed curve shows the calculated transit-broadened resonance which would be expected in the absence of Doppler broadening. The comparison with the observed line shows the strong effect of the time dilation which results in a 25-kHz redshift of the resonance maximum and an additional broadening of almost 100%.

The high stability of the laser offers an easy way to servo lock the laser to the hydrogen transition, for only the slow drift of the reference resonator has to be compensated. For that purpose the computer generates an error signal by subtracting the signal count rate recorded at two different frequencies within the resonance 1 kHz apart. This derivative-shaped signal is translated into a frequency and fed back to the AOM. Figure 3 shows the AOM frequency during 5 min with the servo loop closed. It reflects a linear cavity drift of 1.5 kHz/s but it also can be used to determine the frequency stability of the laser relative to the hydrogen transition. It is better than 1 kHz after an average time of 5 min. This opens the opportunity for an absolute frequency measurement to better than 1 part in 10^{12}. For longer measurement times, like 30 min, another order of magnitude can be expected.

In the near future we will try to resolve the transition to better than 1 kHz. This seems feasible since the current measurement did not exploit the option of our apparatus to cool the atoms to liquid-helium temperature. At 4 K, for example, the Doppler broadening will be reduced by a factor of 50 to 750 Hz. To decrease the transit broadening, apertures can be used to filter out all atoms which do not interact with the optical beam over its entire length of 15 cm. The remaining transit broadening will be about 350 Hz. The related decrease in the count rate is compensated by the longer excitation time resulting in a gain of 75. Another factor of 20 is expected from a more efficient Lyman-α detector using two channeltrons coated with potassium iodide.

The overall linewidth of about 1 kHz can be further reduced by selectively detecting the signal from atoms in the slow tail of the velocity distribution. In time-resolved measurements with the observation period delayed relative to the excitation, we have already observed a significant reduction in the line asymmetry. A time delay of 50 μs is sufficient to reduce second-order Doppler

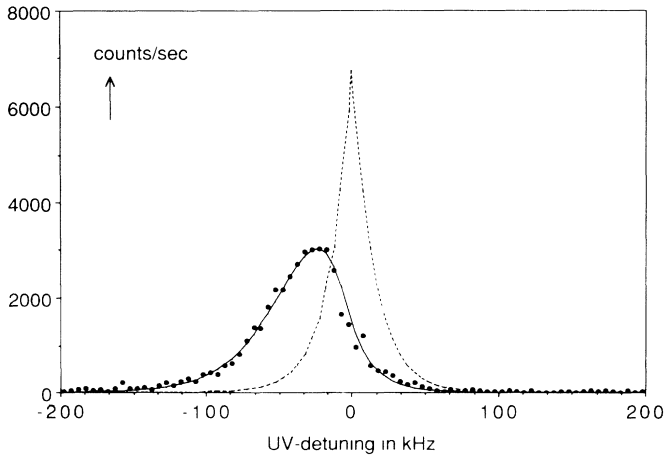

FIG. 2. $1S$-$2S$ two-photon transition ($F=1$). Solid circles: experimental data; solid line: calculated line shape including transit and Doppler broadening; dashed line: transit broadening in the absence of Doppler broadening.

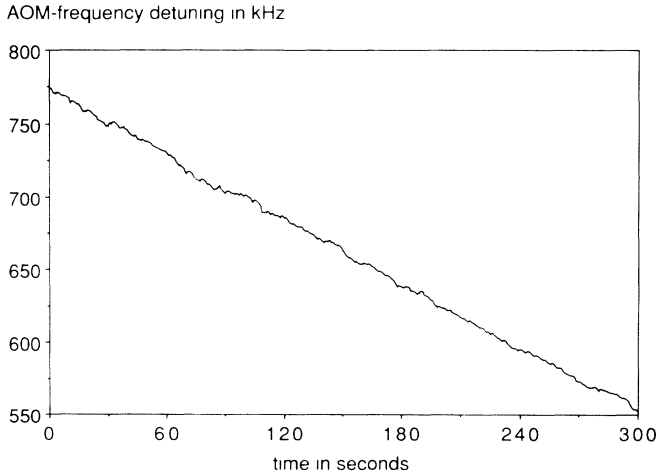

FIG. 3. Frequency stability of the laser servo locked to the $1S$-$2S$ transition: frequency detuning of the acousto-optic modulator during 5 min.

broadening by 50%, while the count rate is reduced to $\frac{1}{5}$. The transit broadening is less affected because it diminishes only with the square root of the temperature. In an experiment at 4 K with a better detector the gain in the count rate of 400 could be used to filter out almost all atoms, keeping only the slowest 0.1%.

At this point it is interesting to compare the potential of an atomic beam to that of a magnetic trap. Since it is possible to confine spin-polarized hydrogen at very low temperatures inside a "magnetic bottle" for long times,[16] spectroscopy on trapped hydrogen atoms could be the next natural step towards resolving the 1-Hz natural linewidth. Zeeman broadening is not as serious as one might at first suspect, since the $1S$-$2S$ transition obeys the selection rule $\Delta F=0$, $\Delta m=0$ and the participating levels are shifted by nearly the same energy. There is, however, a small difference in the g factors of the electron in the two bound states[17] which results in a first-order Zeeman effect of 180 kHz/T. In a trap of about 0.01-T depth, able to contain laser-cooled hydrogen atoms at a few millikelvin, we expect, hence, a Zeeman broadening of several kHz. Unless ways are found to cool the atoms below the recoil limit, or the atoms are released from the perturbing trap in an atomic fountain experiment,[18] a simple beam experiment as reported here will offer an interesting alternative to approach the extremely narrow natural linewidth of the $1S$-$2S$ transition.

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