Measurement of the Hydrogen 1S-2S Transition Frequency by Phase Coherent Comparison with a Microwave Cesium Fountain Clock

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We report on an absolute frequency measurement of the hydrogen 1S-2S two-photon transition in a cold atomic beam with an accuracy of 1.8 parts in 10^{14}. Our experimental result of 2 466 061 413 187 103(46) Hz has been obtained by phase coherent comparison of the hydrogen transition frequency with an atomic cesium fountain clock. Both frequencies are linked with a comb of laser frequencies emitted by a mode locked laser.

The 1S-2S two-photon transition in atomic hydrogen has played a central role in the progress of high resolution laser spectroscopy and optical frequency metrology [1]. It provides a cornerstone for the determination of fundamental constants such as the Rydberg constant, which has become the most precisely measured constant in physics, and the 1S-Lamb shift to yield the most stringent test of quantum electrodynamics in an atom [2–4].

Here, we report on a new measurement of the absolute frequency of the hydrogen 1S-2S transition, which is obtained by phase coherent comparison with an atomic cesium fountain clock as a primary frequency standard. The achieved accuracy of 1.8 parts in 10^{14} surpasses previous measurements of the 1S-2S transition frequency, already the most precisely measured optical transition frequency, by an order of magnitude [5].

The hydrogen 1S-2S transition is excited by Doppler-free two-photon spectroscopy with a standing laser wave near 243 nm in a cold atomic beam. To provide an accurate link between the optical- and radio-frequency domain, we use a novel frequency chain based on measuring absolute frequencies with a train of femtosecond laser pulses [5,6]. The femtosecond laser provides a wide comb of precisely equidistant frequencies, which correspond to the modes of the laser cavity mutually phase locked via Kerr-lense mode locking [7]. The pulse repetition rate, which equals the mode spacing, is electronically phase locked to the radio-frequency standard, such that large optical frequency intervals can be accurately measured. Such a difference frequency measurement can be translated into an absolute frequency measurement by determining the differential frequency between two multiples of an optical frequency. The present measurement takes advantage of a transportable and currently most precise cesium frequency standard in the radio-frequency domain [8,9]. The use of an atomic fountain with laser cooled cesium atoms allows an accuracy of 2×10^{-15}.

At this high level of accuracy, a detailed understanding of the hydrogen 1S-2S two-photon line shapes and several systematic effects, notably the second-order Doppler effect and the ac-Stark shift, becomes necessary. The frequency stability of our chain is sufficient to resolve and explore the ac-Stark shift of this weak UV two-photon transition. With the laser intensities used, this shift amounts to 1 kHz at the most and is thus 2 or 3 orders of magnitude below that observed in hydrogenic 2S-nS and 2S-nD two-photon transitions [3,10]. For an accurate determination of the hydrogen 1S-2S transition frequency, we apply a theoretical model for the line shape taking into account both the excitation geometry and systematic effects [11]. We anticipate that the use of colder hydrogen atoms would allow a considerable further increase in resolution and accuracy.

The achieved accuracy establishes the 1S-2S resonance of atomic hydrogen as a *de facto* optical frequency standard. Ultimately, this hydrogen resonance could allow a definition of time in terms of other fundamental constants. Further, when repeating the frequency measurement with time, the present level of accuracy would already set stringent limits for possible slow relative changes of fundamental constants [12].

The experimental setup is shown in Fig. 1. The hydrogen spectrometer, illustrated at the top of the figure, is similar to the one described previously [11]. The frequency of a dye laser at 486 nm is locked to an external high-finesse reference cavity. The light of this ultrastable laser is frequency doubled, and the resulting UV radiation at 243 nm is coupled into a linear enhancement cavity inside a vacuum chamber for excitation of the two-photon Doppler-free 1S-2S transition. Hydrogen atoms escape from a nozzle cooled to 5–6 K by a liquid-helium flow-through cryostat and form an atomic beam. In the interaction region, shielded from stray electric fields by a Faraday cage, the atoms are excited from the 1S ground state to the excited 2S metastable state. After a distance $d \approx 13$ cm the atoms enter the detection region. A small electric field
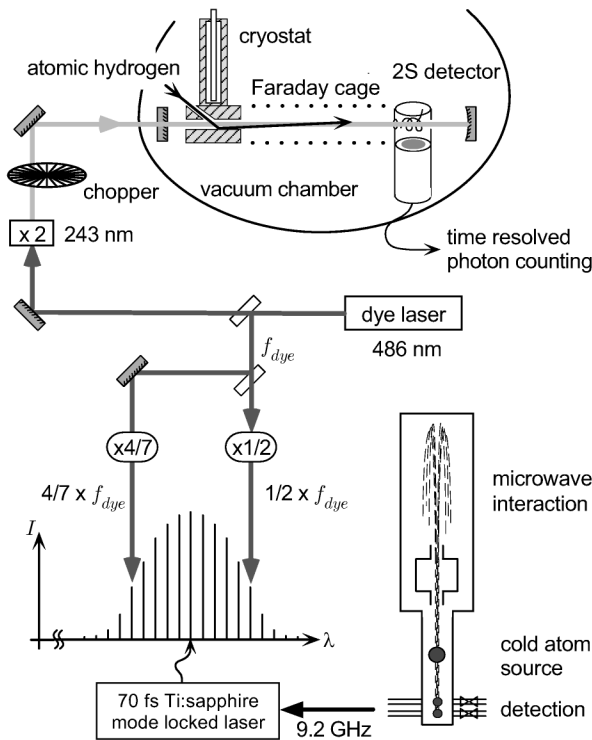
FIG. 1. Experimental setup for comparison of the hydrogen $1S$-$2S$ transition frequency with an atomic cesium fountain clock.

FIG. 2. Time-resolved Doppler-free $1S$-$2S$ two-photon spectrum in atomic hydrogen. The recorded $2S$ count rate is plotted versus delay time and absolute frequency, where $f_0 = 2\,466\,061\,102\,470$ kHz.

mixes the $2S$ and $2P$ states, forcing the excited atoms to decay and emit Lyman-α photons, which can be detected by a photomultiplier. Spurious background counts caused by the excitation light field are efficiently suppressed by periodically blocking the UV light with a mechanical chopper (165 to 465 Hz) and reading out the photomultiplier only in dark times. By enabling signal detection only after a certain delay time τ after blocking the UV light, we select the signal of slow atoms from the thermal beam. This technique further reduces the main systematic effects: the second-order Doppler effect and the time-of-flight broadening. With the help of a multichannel photon counter we detect photons time resolved and record them in a two-dimensional matrix as a function of laser detuning and discrete delay times τ. Figure 2 shows a typical set of 12 simultaneously recorded spectra.

To measure the frequency $f_{1S\text{-}2S}^{\text{hf}}$ of the $1S(F = 1, m_F = \pm 1) \rightarrow 2S(F = 1, m_F = \pm 1)$ transition, we phase coherently compare the frequency of the dye laser $f_{\text{dye}} \approx 616.5$ THz with an atomic cesium fountain clock. As sketched in Fig. 1, the new concept of measuring the frequency of light [5] is based on the measurement of a large frequency difference in the optical region between multiples (harmonics or subharmonics) of a laser frequency. From the dye laser frequency, we first derive the two optical frequencies $4/7 f_{\text{dye}}$ and $1/2 f_{\text{dye}}$ as described below. The frequency gap $4/7 f_{\text{dye}} - 1/2 f_{\text{dye}} = 1/14 f_{\text{dye}} \approx 44$ THz is sufficiently small that it can be measured with the wide comb of regularly spaced laser
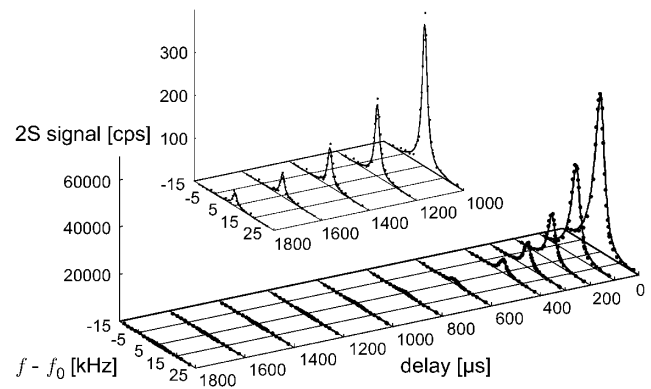
modes emitted by a mode locked femtosecond laser. The separation of these modes is given by the repetition rate of the femtosecond laser, which is phase locked to the radio frequency of the atomic cesium fountain clock.

The subharmonic $1/2 f_{\text{dye}}$ of the dye laser, whose fourth harmonic coincides with the hydrogen $1S$-$2S$ frequency, is produced by locking the second harmonic of a 972 nm laser diode ($2 \times 1/2 f_{\text{dye}}$) to it. Two laser frequencies $1/7 f_{\text{dye}}$ and $4/7 f_{\text{dye}}$ are produced with an optical frequency interval divider [13], which is a means to fix the relation of three laser frequencies such that the center one phase coherently divides the interval spanned by the others: $1/7 f_{\text{dye}}$:$4/7 f_{\text{dye}}$:f_{dye}. At the same time $4/7 f_{\text{dye}}$ is phase locked to the fourth harmonic of $1/7 f_{\text{dye}}$. Because our 3.39 μm He-Ne laser could not be tuned far enough to reach $1/7 f_{\text{dye}}$ we used an additional laser diode and measured a second frequency gap of about 1 THz at its fourth harmonic. A more detailed description of the frequency chain is found in Ref. [5].

The frequency reference for the measurement is provided by a transportable cesium fountain frequency standard, which is described in [9]. A quartz oscillator is frequency locked to the cesium resonance. The relative frequency stability of the device is $1.8 \times 10^{-13}(\text{Hz}\,\tau)^{-1/2}$, where τ is the averaging time, limited by the quartz oscillator phase noise. It has been measured by comparison with a hydrogen maser and corresponds to a relative stability per day of 6×10^{-16}. A preliminary accuracy evaluation of the fountain has been performed. The total uncertainty on the three major frequency shifts, namely, the Zeeman effect, the shift due to blackbody radiation, and the shift due to collisions between cold atoms, is below 10^{-15}. To set an upper limit on other possible frequency shifts, the fountain has been compared to the other BNM-LPTF cesium fountain, FO1, before and after transportation to Garching. Both clocks are found to agree within the 10^{-15} statistical uncertainty of the comparison. A quoted uncertainty of 2×10^{-15} for the absolute frequency of the fountain is then a conservative value.

Figure 2 shows a typical time-resolved hydrogen 1*S*-2*S* two-photon spectrum, as measured by comparison with the cesium atomic clock. We needed about 1 s to record one data point. Within ten days of measurement time we recorded about 600 of such spectra, corresponding to some 2000 data points per day. The high-finesse reference cavity is used as a fly wheel in the optical region. During data analysis, we measure the frequency of the light standing in the cavity and eliminate the drift of our cavity, typically 100 Hz/s at 121 nm, by fitting the drift versus time with a third-order polynomial. The absolute frequency of the dye laser is then calculated from the recorded time tags by adding the frequency difference between the dye laser and the cavity as selected by the acousto-optical modulator. The best fit for the drift is obtained by including not only the time interval for a single recorded spectrum, but an additional 50 data points before and after each spectrum. This procedure is repeated for each spectrum.

To calculate the theoretical 1*S*-2*S* line shape and fit the experimental data, we have numerically integrated the Bloch equations for an atom starting at the nozzle in the 1*S* ground state and traveling on a classical trajectory through the Gaussian 243 nm standing wave to the detector. In addition, we have integrated over all possible trajectories and the atomic velocity distribution $f(v)$. The line-shape model correctly predicts a dependence of the line center frequency on the delay time, as caused by the velocity dependence of the second-order Doppler effect.

A whole set of experimental spectra with different delay times is simultaneously fitted with the Levenberg-Marquart algorithm [14], which is performed with spline interpolated amplitude values from the line-shape model. We use an universal amplitude A and a laser detuning Δ_{det} for all delayed spectra as free parameters. The relative amplitudes and detunings are given by the line-shape model. As the atomic velocity distribution is *a priori* not known with sufficient accuracy, we leave some characteristic parameters of this distribution as additional fitting parameters. The atomic velocity distribution $f(v)$ is given by a Maxwellian velocity distribution [15], which needs to be modified due to the reduction of the number of slow atoms in the atomic beam caused by collisions inside the nozzle, known as the Zacharias effect [16]. The amount of the suppression of slow atoms depends on the Knudsen number $K := \bar{\lambda}/L_n$, where $\bar{\lambda}$ denotes the atomic mean free path length in the nozzle, and L_n is the length of the cylindrical nozzle. In a first step, only the temperature T of the atomic beam and the Knudsen number K were used to describe the atomic velocity distribution. However, satisfactory fits of the time-resolved experimental spectra with the theoretical model could not be obtained until introducing two further fit parameters to more accurately model the atomic velocity distribution. We have accounted for an additional suppression of slow atoms, which we ascribe to atomic collisions with the background gas. This additional suppression is modeled by replacing the v^3 dependence in

$f(v)$ below a certain velocity v_s by $v^{v_{exp}}$. With the two additional fit parameters v_s and v_{exp}, we are able to describe the atomic velocity distribution, as deduced from the time-resolved spectra, quite satisfactorily. To account for additional broadening effects, e.g., the linewidth of the laser, we fold our theoretical spectrum with a Lorentzian profile of linewidth Γ_{Lor} and leave this value as the final fit parameter. The solid lines in Fig. 2 show the result of the described fitting procedure for a typical time-resolved 1*S*-2*S* spectrum.

Besides the second-order Doppler shift, the ac-Stark shift introduces a significant systematic correction to our experimental 1*S*-2*S* two-photon spectra. Our theoretical line-shape model predicts a light shift of 2.45(5) Hz/mW for our excitation geometry when averaging over all possible atomic trajectories. As the shift of the line center is linearly dependent on the excitation light intensity, we can correct for this shift by recording spectra at different light intensities and extrapolating to zero intensity. On a typical measurement day we have recorded 40–90 spectra for different excitation light field powers.

In the upper part of Fig. 3 the measured transition frequencies for a typical day of measurement, as derived from the fits with the theoretical line-shape model, are plotted versus the light power. Linear extrapolation to zero intensity allows the correction for the ac-Stark shift. With this procedure we derive ten values for the 1*S*-2*S* transition frequency and calculate the weighted mean value, as
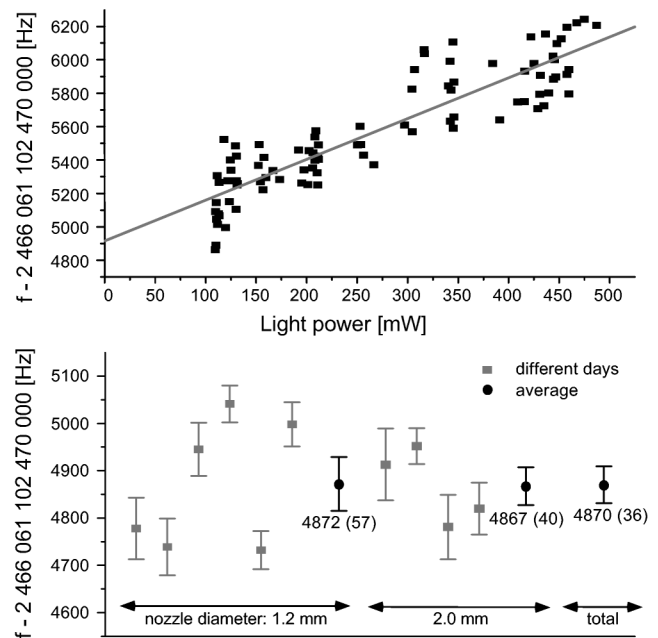

FIG. 3.   Experimental results for the 1*S*-2*S* absolute frequency. The upper graph shows the results for all spectra recorded in a typical measurement day, where the absolute frequency, derived from our line-shape model, is plotted versus the excitation light field power. The graph below shows the complete set of data with the results of extrapolations to zero light intensity for ten different measurement days.

shown in the lower part of Fig. 3. We find a frequency of $f_{1S\text{-}2S}^{\text{hf}} = 2\,466\,061\,102\,474\,870$ Hz, with a statistical uncertainty of 36 Hz. We estimated the systematic uncertainty given by the line shape to be 20 Hz [11], which is mainly determined by the uncertainty to which we can correct for second-order Doppler shift and ac-Stark shift. The slope derived from the linear fit and the measured light power is given by 2.64(45) Hz/mW and is in good agreement with the theoretical value. Line shifts due to blackbody radiation remain below 1 Hz [17].

Residual electric fields lead to a dc-Stark shift of the 1S-2S transition frequency of $\Delta\nu_{\text{dc-Stark}} = 3600E^2$ Hz(V/cm)$^{-2}$ [18]. We estimate stray electric fields to be below 30 mV/cm, corresponding to a dc-Stark effect of less than 5 Hz. The pressure shift of the atomic hydrogen 1S-2S line from molecular hydrogen has been previously measured in a gas cell to be $\Delta\nu_P = 8.4$ MHz/mbar [19]. In our apparatus, the background pressure of 10^{-6} mbar is mainly due to molecular hydrogen, leading to a pressure shift of about 10 Hz. However, the density in the relatively short nozzle region is certainly higher, which could lead to an additional collisional shift of the line center. We have recorded data sets for two different nozzle diameters to vary the pressure inside the nozzle. Figure 3 shows that no statistical significant difference in the results for the line center is observed at the present level of accuracy. Residual recoil shifts due to the finite size of the excitation field optical mode (460 μm diameter) can be estimated to be less than 10 Hz [11].

The frequency $f_{1S\text{-}2S}$ of the hyperfine centroid is obtained by adding a correction $f_{\text{hf}} = 310\,712\,233(13)$ Hz to account for the well known hyperfine splitting of the 1S and 2S levels [20] to the measured transition frequency $f_{1S\text{-}2S}^{\text{hf}}$. Taking the pressure shift into account the obtained result is

$$f_{1S\text{-}2S} = 2\,466\,061\,413\,187\,103(46) \text{ Hz},$$

where the quoted uncertainty was obtained by quadratically adding the uncertainties of the frequencies $f_{1S\text{-}2S}^{\text{hf}}$ and f_{hf}, the systematic uncertainty in the line shape, and 100% uncertainty for the pressure shift, the dc-Stark effect, the blackbody shift, and residual recoil shifts. This result represents the most precisely measured optical frequency in the ultraviolet and visible optical region. Further improvements should be possible by reducing the vacuum background pressure and the stability of the dye laser. Ultimately, a source of colder hydrogen atoms, e.g., in an atomic fountain could allow frequency measurements of the 1S-2S transition with an accuracy exceeding 10^{-16} [21,22].

Both the 1S-Lamb shift and the Rydberg constant can be derived by comparison of two transition frequencies in the hydrogen atom. At the presented level of accuracy, an improvement is here possible only if other hydrogenic transition frequencies, e.g., that of the 2S-nS or 2S-nD ($n = 4, 8, \ldots$) two-photon transitions, are measured more accurately. An interesting option for a comparison with 1S-2S could be the direct excitation of a 1S-nS two-photon line [23].

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