Phase Coherent Vacuum-Ultraviolet to Radio Frequency Comparison with a Mode-Locked Laser

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We demonstrate a versatile new technique that provides a phase coherent link between optical frequencies and the radio frequency domain. The regularly spaced comb of modes of a mode-locked femtosecond laser is used as a precise ruler to measure a large frequency gap between two different multiples (harmonics or subharmonics) of a laser frequency. In this way, we have determined a new value of the hydrogen $1S$-$2S$ two-photon resonance, $f_{1S\text{-}2S} = 2\,466\,061\,413\,187.29(37)$ kHz, representing now the most accurate measurement of an optical frequency.

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Using a new approach to measure the absolute frequency of light with a train of femtosecond laser pulses, we have directly compared the vacuum ultraviolet frequency of the $1S$-$2S$ interval of atomic hydrogen [1] at 2466 THz ($\lambda = 121.6$ nm) with a 10 MHz reference frequency provided by a commercial atomic cesium clock. We reach an accuracy of 1.5 parts in 10^{13}, surpassing the best previous measurement of the $1S$-$2S$ frequency [2], and also a recent record breaking measurement of the Ca intercombination line [3] with a traditional harmonic laser frequency chain [4], thus reestablishing the lead of the $1S$-$2S$ two-photon resonance as the most precisely measured optical frequency in the ultraviolet and visible optical region. A further improvement by an order of magnitude is expected soon when the commercial cesium clock is replaced by a more accurate transportable cesium fountain clock [5].

The hydrogen $1S$-$2S$ resonance with its natural linewidth of only 1 Hz has long inspired advances in high resolution laser spectroscopy and optical frequency metrology, since it provides an important cornerstone for tests of quantum electrodynamic theory (QED), for measurements of fundamental constants, and even for studies of hadronic structure [6]. In the future, it may reveal possible slow changes of fundamental constants or, with the antihydrogen experiments now under preparation at CERN [7], conceivable differences between matter and antimatter.

Recently, we have demonstrated that the femtosecond pulse train of a Kerr-lens mode-locked Ti:sapphire laser can provide a wide comb of precisely equidistant mode frequencies, which can serve as a ruler to measure large optical frequency intervals with unprecedented precision [8,9]. In these experiments, the pulse repetition rate and thus the spacing of the teeth of the frequency comb are electronically phase locked to the radio frequency of a cesium atomic clock which serves as the frequency standard.

Here we demonstrate, for the first time, a new type of laser frequency chain which measures absolute optical frequencies with the help of a femtosecond pulse train. To this end, the interval between two harmonic frequencies of a laser oscillator is bridged with the new frequency ruler. We also take advantage of the precise bisection of optical frequency gaps with an optical interval divider [10,11]. These new tools will soon replace the large, delicate, complex, and highly specialized harmonic laser frequency chains of the past with compact, reliable, and versatile solid state systems for the measurement and synthesis of optical frequencies. We have experimentally demonstrated a potential accuracy of a few parts in 10^{18} [12] for this new type of laser "clockwork," which is likely to play an important role in future generations of atomic clocks that can now be based on sharp optical resonances in atoms [2], ions [13], or molecules.

The reciprocal relationship between time and frequency implies that a single short pulse has a broad spectral bandwidth. A pulse circulating inside the cavity of a mode-locked laser, however, can be described as a superposition of discrete, well-defined laser modes. The resonator provides a long-term phase memory, so that successive emitted pulses have a high degree of mutual phase coherence. The use of such periodic laser pulse trains for the measurement of optical frequency intervals and for high resolution "Ramsey-type" spectroscopy via multiple coherent interactions had been demonstrated in elementary form more than 20 years ago [14]. Similar ideas have also been discussed by Chebotaev [15]. However, phase coherent absolute frequency measurements, as demonstrated here, would not be possible without the more recent dramatic advances in the technology of ultrafast solid state lasers.

Naively, for a laser cavity with dispersive optical elements, one would expect the mode frequencies to be not precisely equally spaced. As a result, a short pulse circulating inside the cavity would tend to spread and change its shape with time. The Kerr lens produced by the circulating light pulse in a nonlinear optical medium via the intensity dependent refractive index counteracts this tendency by periodically modulating the cavity losses. Each mode is, in fact, injection locked by modulation sidebands of the other modes, and will oscillate in precise lock step, as long as the cavity dispersion is sufficiently well

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compensated, so that the mode frequencies do not have to be pulled too far. Modes unable to follow this collective dictate cannot take advantage of the Kerr lens and suffer high round-trip losses in a properly designed cavity.

In the time domain, the output of a mode-locked femtosecond laser may be considered as a continuous carrier wave that is strongly amplitude modulated by a periodic pulse envelope function. If such a pulse train and the light from a cw laser are combined on a photodetector, the beat note between the carrier wave and the cw oscillator is, in fact, observed in a stroboscopic sampling scheme. The detector signal will thus reveal a slow modulation at the beat frequency modulo the sampling rate or pulse repetition frequency.

Of course, the laser spectrum is subject to acoustical and other technical noise. However, in a Kerr-lens mode-locked Ti:sapphire laser, this noise can be readily suppressed with the same servo techniques that are normally used to stabilize the frequency of cw lasers. Carrier frequency and pulse repetition rate, i.e., the position and spacing of the frequency comb, can, in fact, be easily controlled independently by changing the phase and group velocity separately [16]. The ability to measure the absolute frequencies of the laser modes implies that we are now able to measure and stabilize the rate at which the carrier phase slips relative to the pulse envelope [16], or the offset frequency that displaces the laser frequency comb from the harmonic frequencies of the pulse repetition rate. The relative phase between carrier and envelope should become important for nonlinear interactions of light pulses that last for only a few optical cycles.

We were initially surprised that the generation of femtosecond pulses in a Kerr-lens mode-locked laser is so highly reproducible that the spectral modes remain precisely equidistant out into the far wings of the spectrum. We have since shown that this remains true even if the laser spectrum is further broadened by self phase modulation in a nonlinear optical fiber [12]. Recently, extreme spectral broadening has been observed with "photonic crystal fibers" [17], which manage the optical dispersion with the help of a microstructured cladding. Meanwhile we have shown [12] that such highly nonlinear fibers can preserve the phase coherence between successive pulses, so that frequency combs spanning more than one octave are becoming available. A compact frequency chain consisting of two lasers only (a frequency doubled Nd:YAG and a femtosecond laser) has already been operated in our laboratory [12]. A high degree of mutual phase coherence has even been observed for pairs of white light continuum pulses produced by focusing the light from an amplified Ti:sapphire femtosecond laser at two separate spots inside a nonlinear medium, despite complications such as self-focusing, stimulated Raman and Brillouin scattering, or shock wave formation [18]. Similar experiments with high harmonics produced in gas jets [19] suggest that it may eventually become feasible to extend absolute optical frequency measurements into the extreme ultraviolet and soft x-ray region.

To demonstrate the new approach to optical frequency metrology in the most expeditious way, we start with an existing laser frequency chain, as described in detail elsewhere [2], and we introduce a commercial Kerr-lens mode-locked Ti:sapphire laser (Coherent, model Mira 900) as an active frequency comb generator to measure two large optical frequency gaps as illustrated in Fig. 1.

Previously, the 3.39 μm CH_4-stabilized He-Ne laser with its frequency $f \approx 88.4$ THz served as the reference standard. In the new chain, the He-Ne laser acts only as an optical flywheel. The reference standard is now a cesium atomic clock (Hewlett Packard, model 5071A), which controls the repetition rate of the femtosecond laser and thus all the optical frequencies in the chain.

The dye laser for the hydrogen spectrometer operates at a frequency $7f - 2\Delta f$, where the offset $\Delta f \approx 1$ THz posed a major challenge in earlier experiments [2]. As before, we produce the fourth harmonic $4f$ of the He-Ne laser in two frequency doubling steps, using a phase-locked 1.70 μm NaCl:OH^- color center laser at $2f$ and an 848 nm diode laser at $4f$ as transfer oscillators (not shown in Fig. 1). An optical frequency interval divider [11] between f and $7f - 2\Delta f$ generates the precise center frequency $4f - \Delta f$ (oval symbol in Fig. 1). The relatively small interval Δf between $4f$ and $4f - \Delta f$ can now be measured effortlessly with the much broader frequency comb of the mode-locked femtosecond laser.

To measure the absolute optical frequencies, we introduce a 973 nm diode laser whose second harmonic frequency (generated in a B-cut $KNbO_3$ crystal) is phase

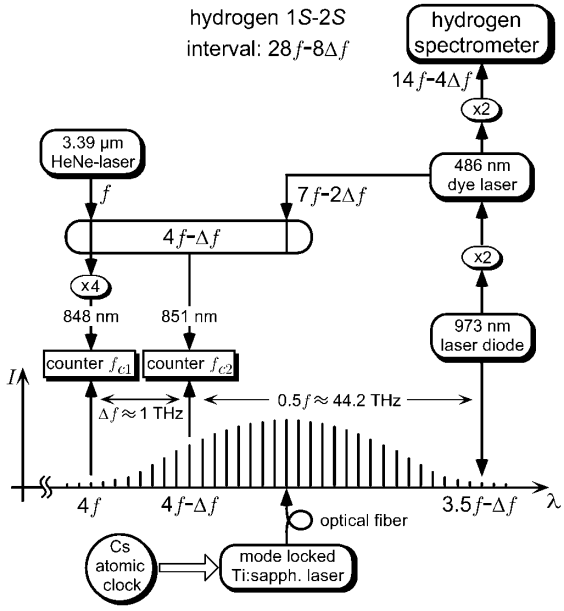

FIG. 1. Novel frequency chain for the measurement of the hydrogen 1S-2S interval. The frequency comb of a mode-locked laser is used to measure two large optical frequency differences.

locked to the dye laser, so that it oscillates at the precise subharmonic $3.5f - \Delta f$. By bridging the interval of $0.5f$ between $4f - \Delta f$ and $3.5f - \Delta f$ with the frequency comb of the femtosecond laser, we are, in effect, measuring the frequency f of the He-Ne laser directly in terms of the atomic clock frequency. Since the Mira laser has a rather moderate pulse length of 73 fs, we send its output through a 40 cm long single mode fiber which broadens the spectrum by self-phase modulation so that it bridges the interval of 45.2 THz [12,16].

A single step link between the 45.2 THz interval and the 10 MHz output frequency of the cesium clock is established by stabilizing the 200th harmonic of the pulse repetition rate to a 15.2 GHz signal [16] provided by a microwave synthesizer (Hewlett Packard, model 83623A). The synthesizer is referenced to the cesium clock, which is continuously monitored using a global positioning system (GPS) time receiver (Hameg, model 8125). In addition to the mode spacing, we "fix" the entire comb by stabilizing the beat signal between the dye laser and the second harmonic of the 973 nm laser diode, which is phase locked to a particular mode at $3.5f - \Delta f$. The simultaneous control of the mode spacing and the absolute position of our comparatively narrow frequency comb [16] already enables us to measure any optical frequency between 848 and 973 nm.

All frequency locks in the chain are optical phase-locked loops [20], which control the frequency of one laser, so that the beat frequency with the second laser oscillates in phase with a reference (or local oscillator) frequency provided by our cesium atomic clock. To continuously monitor the performance of these servo loops, we count all in-lock beat signals with radio frequency counters (Hewlett Packard, model HP 53131A) using a different filter bandwidth as used for the phase detectors. In this way, possible cycle slipping events are effectively removed from our data [9].

As shown in Fig. 1, we measure the beat frequencies f_{c1} and f_{c2} at $4f$ and $4f - \Delta f$ with the modes of the comb. To ensure that the beat notes are counted correctly, we use additional laser diodes (not shown in Fig. 1) phase locked to these modes to provide enough power for a strong beat signal (signal to noise ratio $\geq$40 dB in 400 kHz detection bandwidth). In the phase-locked condition, the chain relates the frequency of the hydrogen $1S$-$2S$ transition ($28f - 8\Delta f$) to the measured radio frequencies f_{c1} and f_{c2} through

$$\begin{aligned} f_{1S\text{-}2S} &= -8f_{c1} - 64f_{c2} + (-8N_1 + 64N_2)f_r + f_{\mathrm{LO}} \\ &= -8f_{c1} - 64f_{c2} + 2466.063\,84\ \mathrm{THz}, \end{aligned} \tag{1}$$

where $N_1 = 595\,351$ and $N_2 = 581\,421$ are the number of modes between the 973 nm laser diode and the modes that produce the measured beat notes, $f_r = 76$ MHz is the pulse repetition rate, and $f_{\mathrm{LO}} = 5.504$ GHz contains all contributions from the local oscillator frequencies used for phase locking.

The hydrogen spectrometer has been described in detail previously [21]. The extremely narrow $1S_{1/2}(F = 1, m_F = \pm 1) \rightarrow 2S_{1/2}(F' = 1, m_{F'} = \pm 1)$ Doppler-free two-photon resonance at 243 nm is driven in a cold atomic beam by a frequency doubled ultrastable dye laser. The UV radiation is resonantly enhanced in a linear cavity inside a vacuum chamber and excites the atoms traveling collinear to this standing wave. At a distance of $l \approx 14$ cm downstream from the beam nozzle atoms that have been excited to the metastable $2S$ state are probed with the help of a quenching electric field that forces the emission of Lyman-α photons. In order to reduce time-of-flight broadening and second order Doppler shifts, we select slow atoms from the thermal beam by turning the laser light periodically off with a chopper and counting signal photons only if they arrive after a time delay of $\tau = 1.2$ ms.

We determine the center of the recorded lines by fitting a Lorentzian to the experimental data. As confirmed by simulations [22], this procedure is sufficiently precise at present if the second order Doppler shift is corrected appropriately. An upper limit for the second order Doppler shift $\Delta\nu = -1/2\ (v/c)^2 f_{1S\text{-}2S}$ is given by $v = v_{\max} = l/\tau$. A detailed model for the line shape of the resonance [22], however, predicts that for sufficiently long delay times an effective velocity $\bar{v} \approx v_{\max}/2$ gives a realistic estimate for the second order Doppler shift, as the slowest atoms spend the longest time for excitation in the light field. Assuming an uncertainty of 50% in $\bar{v}$, we correct for a redshift of $1.9 \times 10^{-14} \pm 1.9 \times 10^{-14}$. Averaging the actual particle trajectories in the laser light field, the line shape model [22] yields an ac Stark blueshift of 2.45(5) Hz/mW, corresponding to a correction of $-2.7 \times 10^{-13} \pm 1.3 \times 10^{-13}$. The relative uncertainty of 50% in the (linear power-dependent) ac Stark shift is due to a 50% uncertainty in the absolute determination of the light power inside the enhancement cavity. Other uncertainties are estimated to contribute less than 8×10^{-15} [22,23].

During the experiment the dye laser is locked to a stable Zerodur cavity. An acousto-optic modulator (AOM) is used to shift the dye laser frequency relative to the frequency of the light in the cavity, so that the dye laser can be scanned over the atomic resonance. For the data analysis, we first determine the frequency of the light in the cavity with our frequency chain with a counter gate time of one second. The cavity has a superior short term stability so that the phase stability of the frequency chain including its cesium reference (specified Allan standard deviation 5×10^{-12} within one second) can be derived. The measured scatter of cavity frequency readings was reduced by a factor of 10 (to the specified Allan standard deviation of 5×10^{-13} within one second) when we replaced the quartz oscillator at the output of the cesium clock by a quartz with a tenfold improved stability. Thus, with the present radio frequency reference, the frequency chain contributes at a negligible level to the total measurement noise.

To use the cavity, that is drifting at a relative rate of up to 6×10^{-14} per second, as a flywheel in the optical region we fit a third order polynomial to its frequency as a function of time (for periods of usually more than 3 h). This polynomial is used to calculate the absolute frequencies of the $1S$-$2S$ line centers from the recorded number of emitted Lyman-α photons, the AOM operating frequency, and the recorded time tags. For a typical day of measurement, we obtain about 35 recorded hydrogen spectra and a total of 900 s of measurement data valid for the frequency evaluation of the reference cavity.

Evaluating the data of ten days of measurement, we determine the frequency of the $1S_{1/2}(F = 1) \rightarrow 2S_{1/2}(F' = 1)$ transition as the weighted mean value of the daily results. The scatter of the one-day mean values of the line centers is due to two statistical effects: the cesium clock instability of 1.5×10^{-13} (specified to less than 2.8×10^{-13}) within 900 s and the statistical uncertainty of around 10^{-13} due to the fitting procedures (polynomials and Lorentzians). For the full ten days of data acquisition, this reduces to a total statistical uncertainty of 5.7×10^{-14}.

We further apply corrections due to small deviations of the GPS time from the universal time, as published by the Bureau International des Poids et Mesures in Paris. In addition, we correct for the gravitational redshift at the site of our laboratory at 490 m above sea level by subtracting 5.3×10^{-14} from the result. We conservatively estimate the uncertainty of this calibration of our cesium clock to be less than 2 parts in 10^{14}.

To account for the well-known hyperfine splittings of the $1S$ and $2S$ levels [24], we add 310 712 223(13) Hz to the result to obtain the $1S$-$2S$ hyperfine centroid frequency:

$$f_{1S\text{-}2S} = 2\,466\,061\,413\,187.29(37)\ \text{kHz}\,. \qquad (2)$$

The error budget is dominated by the 1.3×10^{-13} uncertainty of the ac Stark shift and the 5.7×10^{-14} statistical uncertainty of the mean value of the line centers. Both contributions can be reduced significantly if a more stable radio frequency source is used. In this case measurements with different excitation powers can be performed in a reasonable integration time to extrapolate to zero ac Stark shift.

In conclusion, we have performed the first phase coherent radio frequency to vacuum UV comparison to measure the $1S$-$2S$ transition frequency of atomic hydrogen. The measurement is limited by the short-term stability of our local cesium atomic clock. Together with a new, more accurate value of the proton charge radius [25] and planned precise absolute frequency measurements of hydrogen transitions between excited states with ultracold atoms [26], our measurement will allow more stringent tests of bound-state QED theory.

Even though the reported experiments are based on an existing complex harmonic frequency chain, the techniques introduced here can be applied with reliable and much more compact solid state and semiconductor lasers. This opens for the first time the possibility to construct an optical clock with a clockwork that can continuously work for an extended period of time.

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