Ultraprecise Measurement of Optical Frequency Ratios

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We developed a novel technique for frequency measurement and synthesis, based on the operation of a femtosecond comb generator as transfer oscillator. The technique can be used to measure frequency ratios of any optical signals throughout the visible and near-infrared part of the spectrum. Relative uncertainties of 10^{-18} for averaging times of 100 s are possible. Using a Nd:YAG laser in combination with a nonlinear crystal we measured the frequency ratio of the second harmonic ν_{SH} at 532 nm to the fundamental ν_0 at 1064 nm, $\nu_{SH}/\nu_0 = 2.000\,000\,000\,000\,000\,001 \times (1 \pm 7 \times 10^{-19})$.

The development of optical frequency comb generators based on Kerr-lens mode-locked femtosecond lasers [1,2] has enormously stimulated the field of optical frequency synthesis and metrology. Using this technique, the absolute frequencies of a number of narrow transitions in cold atoms or single stored ions such as H, Ca, Yb^+, or Hg^+ have been measured by phase-coherently linking those signals to primary cesium-clock controlled hydrogen masers [3–6]. The measurement instabilities approached those of the hydrogen masers, indicating that neither the optical frequency standards nor the frequency combs themselves were limiting parts of the setups.

Any absolute frequency measurement is finally limited by the frequency instability of the device realizing the unit of frequency, Hertz, such as a radio or microwave reference like the hydrogen maser. A possibility to avoid this limitation is the measurement of optical frequency *ratios,* which are unitless. Thus, frequency ratios for oscillators with better stability than that of the radio or microwave reference can be determined with smaller uncertainty than the absolute frequencies if a technique is available to realize the frequency ratio without introducing additional noise.

Such a technique is the transfer oscillator concept, which has been realized with a harmonic frequency chain [7]. However, the measured frequency ratios were restricted to small integer numbers. In this Letter, we describe a novel technique based on the operation of a femtosecond frequency comb generator as a transfer oscillator. Our technique has the capability of generating *arbitrary* ratios of any optical frequencies throughout the visible and near-infrared part of the spectrum, while frequency fluctuations of the comb modes do not enter the measurement but cancel out.

We demonstrate the superior short-term instability by two measurements: first, we measured the frequency ratio of signals from a single-Yb^+-ion frequency standard [8] and from an I_2-frequency-stabilized Nd:YAG laser (yttrium-aluminum-garnet) [9]. Second, we used the Nd:YAG laser and measured the frequency ratio of the second harmonic at 532 nm to its fundamental at 1064 nm, thereby testing how accurately the 2:1 frequency ratio

is realized by second harmonic generation. We demonstrate the capability of our technique of frequency-ratio measurements with relative uncertainty better than 10^{-18}.

Kerr-lens mode-locked femtosecond lasers emit a periodic train of short pulses. The spectrum of this emission corresponds to a comb of distinct modes, that are exactly equally spaced due to the strong and fast Kerr-lens mode-coupling mechanism [1]. Any external signal with frequency ν_i within the comb spectrum can be related to a suitable comb mode by detection of the beat-note frequency Δ_i,

$$\nu_i(t) = \nu_{ceo}(t) + m_i f_{rep}(t) + \Delta_i(t). \qquad (1)$$

Here, m_i denotes the integer order number of the comb mode selected for the beat note, f_{rep} the pulse repetition rate, and ν_{ceo} a so-called carrier-envelope offset frequency that accounts for the offset of the entire comb with respect to the frequency zero. For absolute frequency measurements all detected frequencies, ν_{ceo}, f_{rep}, and Δ_i are referred to a microwave-reference frequency f_R, which may be generated by a primary clock.

The task of optical frequency metrology in general is the establishment of a phase-coherent link between two or more external frequencies $f_R, \nu_1, \nu_2, \ldots,$ while the noise contributions from the frequency link, such as the femtosecond comb's parameters ν_{ceo} and f_{rep}, are kept as small as possible. Conventional approaches [10,11] aim to reduce fluctuations of ν_{ceo} and f_{rep} by stabilization of both the group and phase delay of the laser resonator, e.g., with the help of piezo transducers. However, as a result of the finite response time of these elements, the servo bandwidth of such servo loops is generally not sufficient to reduce the frequency noise of the beat notes Δ_i to that of the optical signals at ν_i. The stability of f_{rep} is particularly demanding, since according to Eq. (1) this quantity is multiplied by the mode number m_i, which is of the order of 10^6. As a consequence, the short-term instability of such frequency measurements may be limited by noise from the microwave reference imposed on the frequency comb. Nevertheless, a remarkably low instability

of 7×10^{-15} at 1 s has been achieved with this technique [11].

Our novel approach completely differs in handling the technical frequency contributions. We synthesize one radio frequency signal containing all necessary information from the external optical signals. This radio frequency is independent of ν_{ceo} and f_{rep} and is thus immune against noise contributions from the femtosecond laser. The frequency ν_{ceo} cancels out in real-time signal processing and the detection of f_{rep} is not required at all.

The setup is sketched in Fig. 1. Three signals are detected: ν_{ceo} and two beat signals of two external frequency standards with modes of the femtosecond frequency comb, Δ_1 and Δ_2. The sum frequency of Δ_1 and ν_{ceo} is generated by a mixer. The sum frequency of Δ_2 and ν_{ceo} is additionally processed with a direct-digital-synthesis IC (DDS). Such a device generates an output signal from an input signal with a frequency ratio given by a long digital tuning word. It numerically approximates the ratio of the two integers m_1 and m_2 by $j/2^n$, where j is an integer and n the bit length of the tuning word. The resulting possible error is negligible for commercially available $n = 48$ bit devices. As a result of this signal processing, one obtains

$$\nu_A = \nu_{\text{ceo}} + \Delta_1$$

and

$$\nu_B = (\nu_{\text{ceo}} + \Delta_2) \frac{m_1}{m_2}.$$

Generating the difference frequency of both signals with the help of a mixer, we find using Eq. (1)

$$\nu_C = \nu_A - \nu_B = \nu_1 - \nu_2 \frac{m_1}{m_2}.$$

This frequency ν_C is a measure for the (small) deviation of the optical signal's frequency ratio ν_1/ν_2 from m_1/m_2. Since $\nu_C \ll \nu_1, \nu_2$, the requirements on the radio frequency reference are in general not demanding, i.e., it is not necessary to refer ν_C to a cesium-clock controlled hydrogen maser. Alternatively, ν_C can be referred to one of the optical frequency standards with the help of the detection of the repetition rate f_{rep}. In this case, a self-referenced measurement system is established, independent of the realization of the unit Hertz.

The frequency ν_C carries the full information of the fluctuations of the ratio ν_1/ν_2, but it is independent of the properties of the femtosecond laser, i.e., it is independent of both the frequency fluctuations of ν_{ceo} and the repetition rate f_{rep}. The complete cancellation of all femtosecond comb parameters realizes the transfer principle. The essential requirement is that Eq. (1) holds for the instantaneous phase angles. Technically, the cancellation requires a sufficiently fast detection of the optical signals, but no active stabilization as in the conventional approach mentioned above. This requirement can be easily fulfilled using fast photodetectors given the typical Fourier frequency range of the noise of ν_{ceo} and the beat signals Δ_1 and Δ_2.

The femtosecond frequency comb is generated by our Kerr-lens mode-locked Ti:sapphire laser, similar to that described in [12]. The pulse duration and repetition rate are <15 fs (FWHM) and 100 MHz, respectively. Approximately 30 mW of the laser output is coupled into a 10 cm long piece of air-silica microstructure (MS) fiber [13], leading to an output spectrum that extends from about 500 nm to about 1100 nm. The carrier-envelope-offset frequency ν_{ceo} is measured via the beat note of a few ten thousand frequency-doubled infrared comb lines at 1080 nm with fundamental lines at 540 nm. The beat notes of the external optical signals with comb modes are detected with fast pin photo diodes. More technical details related to the setup and the signal processing scheme in Fig. 1 can be found in Ref. [14].

For the two frequency ratio measurements we chose two frequency standards generating three different reference frequencies: (i) The subharmonic of the output of a single-ytterbium-ion frequency standard [8] at $\nu_{\text{Yb}} = 344\,179\,449$ MHz (871 nm), (ii) the single-frequency output of a Nd:YAG laser at $\nu_0 = 281\,606\,335$ MHz (1064 nm), that was frequency stabilized to an iodine transition, and (iii) the second harmonic of the Nd:YAG laser's emission at $\nu_{\text{SH}} = 563\,212\,670$ MHz (532 nm), generated by a periodically poled KTP crystal.

Both the frequency ν_{Yb} [6] and ν_0 [15] were previously measured with respect to a primary cesium atomic clock.

In the first experiment we measured the ratio between ν_{Yb} and ν_0. With respect to Fig. 1, the beat note of ν_{Yb} with a comb mode was Δ_1 and that of ν_0 was Δ_2. We obtained

$$\frac{\nu_{\text{Yb}}}{\nu_0} = 1.222\,200\,739\,113 \times (1 \pm 3 \times 10^{-12}).$$

The uncertainty was dominated by the reproducibility of the iodine-stabilized Nd:YAG laser.

The Allan standard deviation is shown in Fig. 2 (squares). The data for short averaging times (open
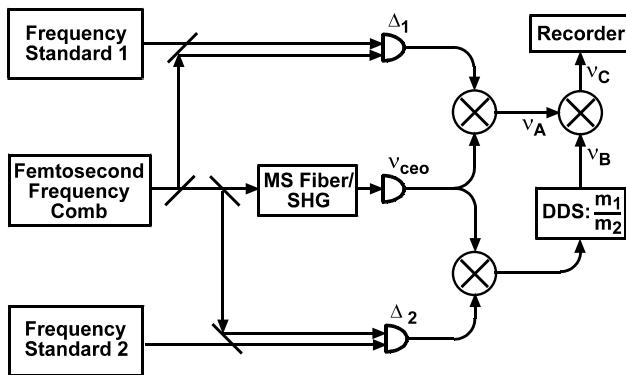

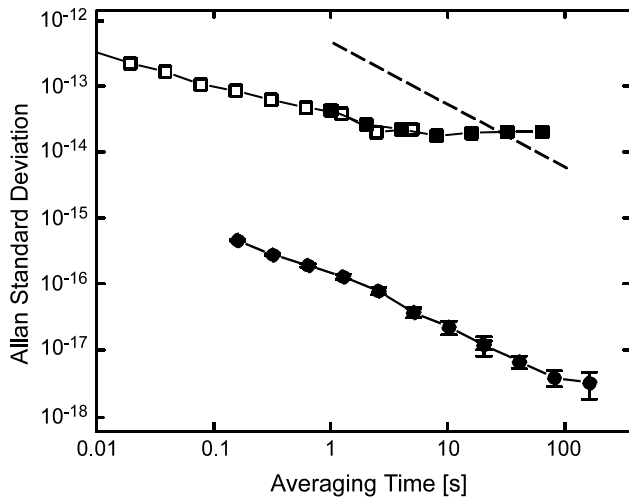FIG. 1. Linking two optical frequencies with a femtosecond comb: real-time signal processing scheme.

FIG. 2.   Measured Allan standard deviation of two frequency-ratio measurements using the Yb^+ and Nd:YAG reference frequencies (squares) and the second harmonic and fundamental of a Nd:YAG laser (circles). The frequency instability of a typical microwave reference (hydrogen maser) is included for comparison (dashed line).

squares) were derived from the signal ν_c (in Fig. 1). For long averaging times, the real-time processing is not necessarily required. Instead, the frequency ratio can be calculated offline from averaged values (e.g., 1 s) of ν_{ceo}, Δ_1, and Δ_2 (solid squares in Fig. 2). Both data sets agree in the overlapping range between 1 and 8 s.

Similar uncertainty levels have been obtained by direct comparison of two Nd:YAG lasers, indicating a limitation of our measurement by the Nd:YAG laser. We concluded that the stability of the reference signal pair was not good enough to investigate any possible noise contribution from the femtosecond comb. Therefore we used in the second experiment the signal ν_0 and its second harmonic ν_{SH} that was generated in a nonlinear crystal. The frequency-doubling process enabled the generation of a pair of reference frequencies with completely correlated fluctuations, leading to a fixed frequency ratio 2:1 even for the shortest averaging times. With respect to Fig. 1, the beat note of ν_{SH} with a comb mode was Δ_1 and that of ν_0 was Δ_2. The DDS was replaced by a frequency doubler. Both comb lines generating the beat notes at the fundamental and the second harmonic were themselves generated in the microstructure fiber.

The experimental result for the frequency ratio of the second harmonic and the fundamental from the Nd:YAG laser is

$$\frac{\nu_{SH}}{\nu_0} = 2.000\,000\,000\,000\,000\,001 \times (1 \pm 7 \times 10^{-19}).$$

This measurement sets a new limit on the relative error of the 1:2 ratio of the frequency doubling process in a nonlinear crystal of smaller than 10^{-18}. This limit is more than 5 orders of magnitude below previous experimental limits [16]. It should be noted that second harmonic gen-

eration has been employed not only for the generation of ν_{SH} but also for the generation of ν_{ceo}. However, the conditions differed substantially. Whereas ν_{ceo} was generated by SHG of many thousands of comb line near 1080 nm, ν_{SH} was obtained by frequency doubling of one single line.

Figure 2 shows the Allan standard deviation for the measurement (circles). A value of 1×10^{-16} is achieved at 1 s. Indeed, the instability is more than 2 orders of magnitude smaller than for the ν_{Yb}/ν_0 measurement, for which already high quality oscillators were used, and it surpasses even the performance of state-of-the-art optical oscillators [17] substantially. The observed stability can be explained by the estimated fluctuations of the length difference between the beam paths of the two optical signals. Thus one might expect an even better stability when path length fluctuations are technically suppressed. Such fluctuations usually exhibit complicated spectra, thus explaining why the shape of the Allan standard deviation in Fig. 2 differs from the $1/\tau$ dependence that would be expected for phase-coherent processes with white phase noise level.

A quantum noise contribution to the short-term instability is not yet significant. If we assume that the signals of both optical clocks, that are to be compared, can be phase tracked with a bandwidth of 1 kHz, we estimate from our experimentally observed, shot-noise limited signal-to-noise ratio an Allan standard deviation of 3×10^{-18} s$/\tau$, which is more than 1 order of magnitude smaller than the values of Fig. 2.

A typical record of the phase of the signal ν_C in Fig. 1 is shown in Fig. 3. It was measured with respect to a fixed 25 Hz offset signal. The phase excursions were smaller than 1 rad for periods of minutes. From such phase records we obtained by fast Fourier transform a line width of 9 mHz, as shown in Fig. 4. It should be noted that this line was derived from beat notes Δ_1 and Δ_2 with common mode frequency excursions as wide as MHz.
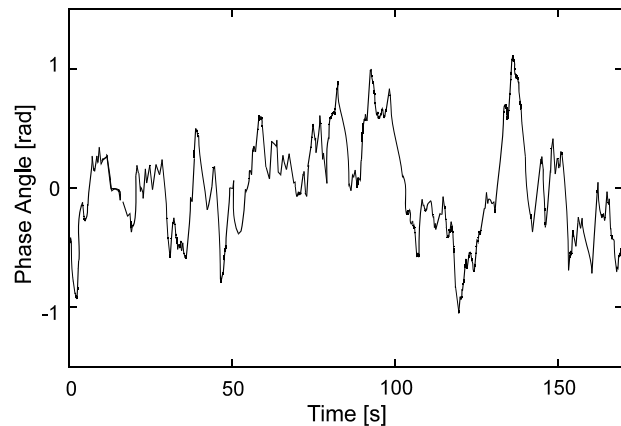

FIG. 3.   Typical relative phase excursions of ν_C (see Fig. 1), representing the processed second harmonic and fundamental of the Nd:YAG laser. In order to avoid base-band detection, the frequencies ν_A and ν_B were frequency shifted leading to an average value of 25 Hz (arbitrarily chosen) of ν_C.
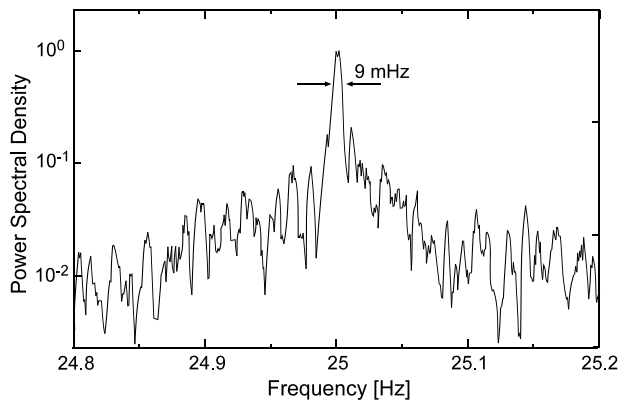
FIG. 4.   Spectrum of ν_C, calculated from the phase excursions in Fig. 3 by fast Fourier transform. A 900 s data string was used for the calculation.

In conclusion, we have demonstrated a novel concept for frequency measurement and synthesis, based on the operation of a femtosecond comb generator as transfer oscillator, which does not introduce additional noise. It allows one to exploit the superior short-term stability of optical frequency standards by the measurement of frequency ratios. Indeed, the transfer principle using a femtosecond comb is not limited to optical frequencies, but can also be applied to radio and microwave frequencies [14]. The capability of such high precision measurements enables new applications in spectroscopy as soon as appropriate reference signals from optical transitions can be realized with sufficient frequency stability. Interesting questions are effects such as QED corrections, isotope shifts, or relativistic corrections to absolute transition frequencies [18]. Related to the expanding Universe, a variation of fundamental constants such as the coupling strengths of electromagnetic, weak, or strong interaction is expected. These constants are indeed accessible with optical spectroscopy [19]. Relative frequency measurements of optical transitions with different scaling in the fine structure constant α are sensitive to temporal changes of the electromagnetic coupling strength. Comparison of hyperfine splitting provides an additional sensitivity to changes of the strong interaction strength. So far, laboratory experiments have set limits on the variations of about 10^{-14} yr^{-1}. Our frequency-ratio measurement technique, which is capable of linking optical signals with an uncertainty of 10^{-18} in 100 s, may be part of substantially improved experiments.

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