

Phase-coherent frequency measurement of the Ca intercombination line at 657 nm with a Kerr-lens mode-locked femtosecond laser

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The frequency of the calcium $^3P_1-^1S_0$ intercombination line at 657 nm is phase-coherently measured in terms of the output of a primary cesium frequency standard using an optical frequency comb generator comprising a sub-10-fs Kerr-lens mode-locked Ti:sapphire laser and an external microstructure fiber for self-phase-modulation. The measured frequency of $\nu_{\text{Ca}}=455\,986\,240\,494\,276$ Hz agrees within its relative uncertainty of 4×10^{-13} with the values previously measured with a conceptually different harmonic frequency chain and with the value recommended for the realization of the SI unit of length.

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So far, the use of optical frequency standards for the realization of basic units, e.g., of time and length, was hampered by the difficulties of measuring their high frequencies. As a result, the most precise frequency values recommended by the Comité International des Poids et Mesures (CIPM) are based on very few frequency measurements [1]. Recent progress in femtosecond pulse generation and the advent of microstructure optical fibers [2–4], however, have substantially facilitated such optical frequency measurements. These schemes [5–7] start with the highly periodic pulse train of a Kerr-lens mode-locked laser that corresponds in the frequency domain to a comblike frequency spectrum of equidistant lines. The spectral span of this comb reflects the duration of an individual pulse, while the spacing between the lines is determined by the pulse repetition frequency. It has been shown that the fast, spectrally far-reaching Kerr-lens mode-locking mechanism enforces a tight coupling [8] of the optical phases of the individual lines. As a result, the frequency of any of these lines is given by an integer multiple of the pulse repetition frequency f_{rep} and a frequency ν_{ceo} [9–11] that accounts for the offset of the entire comb with respect to the frequency origin, e.g., for the n th mode $\nu_n = \nu_{\text{ceo}} + n f_{\text{rep}}$. If these quantities are known, the unknown frequency of any external optical signal can be precisely determined by counting the frequency of the beat note between this signal and a suitable comb line. In this Rapid Communication, we report an application of this method to phase-coherent measurements of the frequency of an optical Ca frequency standard operating on the $^1S_0-^3P_1$ intercombination transition at 657 nm.

The frequency of this state-of-the-art standard has already been measured with a conventional harmonic frequency chain [12,13], and the frequency value of $\nu_{\text{Ca}} = 455\,986\,240\,494\,150$ Hz has been recommended by the CIPM for the ‘‘Practical realization of the definition of the meter (1997)’’ [1] with a relative standard uncertainty of 6×10^{-13} . The present experiments allow the comparison of two conceptually different phase-coherent optical frequency measurement concepts, the conventional type operating in

the absolute-frequency domain and the novel type operating in the difference-frequency domain. This comparison is performed without the need for intermediate transportable standards.

Two of the three required measurements mentioned above, the pulse repetition rate f_{rep} and its multiple integer factor, are straightforward to carry out. The measurement of the carrier-envelope offset frequency ν_{ceo} , which arises from the relative velocity of the carrier phase and the pulse envelope, is a more demanding task. Several more or less complex schemes, depending on the comb span available, have been proposed for the measurement of ν_{ceo} [10]. The simplest concept requires an octave span of the frequency comb that is not directly available from our laser. Thus, the available span has to be expanded, e.g., by external self-phase modulation (SPM). Microstructure air-silica fibers, which allow for tailoring the group-velocity dispersion (GVD) properties [2] are highly suited for SPM of moderate peak power pulses available from mode-locked laser oscillators since they provide both lateral and temporal confinement of the pulses over long interaction lengths. If the octave span is achieved, then the ν_{ceo} measurement can be accomplished by second-harmonic generation (SHG) of the spectral lines in the low-frequency wing of the comb. Whereas the frequencies of the comb lines are shifted by ν_{ceo} with respect to the origin, their harmonics are shifted by $2\nu_{\text{ceo}}$. Thus, the beat notes between these harmonics and the corresponding high-frequency-wing lines of the comb show the desired component at ν_{ceo} .

As depicted in Fig. 1, our measurement setup consists of four parts: the frequency comb generator (i), the detectors and electronics for servo control and/or data acquisition of the pulse repetition frequency (ii), the beat note between the laser and Ca-standard radiation (iii), and the carrier-envelope-offset frequency (iv).

(i) The Kerr-lens mode-locked (KLM) Ti:sapphire laser is similar to that described in [14]. The laser generates pulses with a duration of ~ 10 fs, a repetition rate $f_{\text{rep}} = 100$ MHz, an average power of 120 mW, and a spectrum centered at

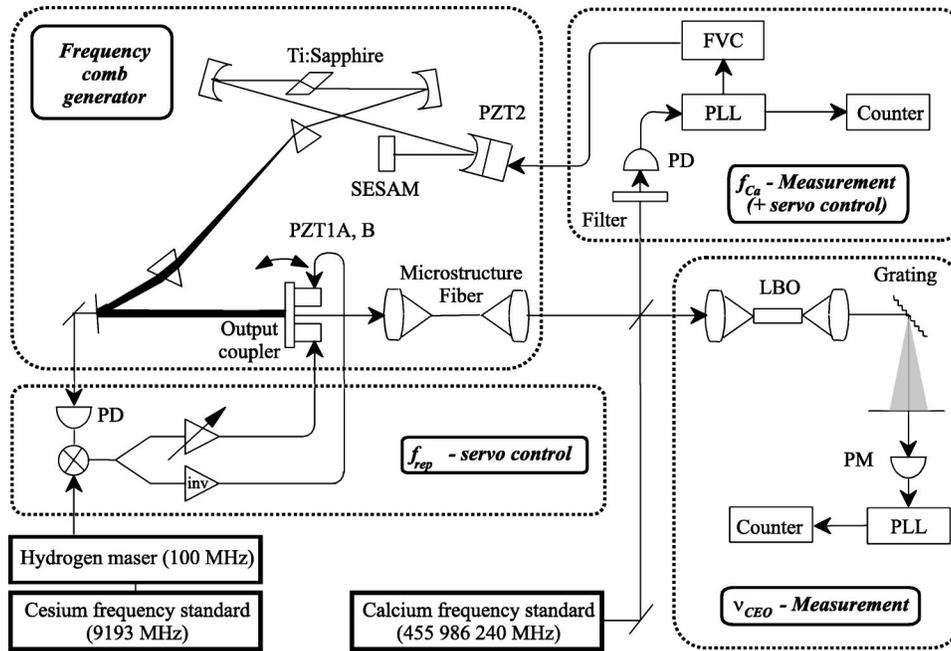


FIG. 1. Schematic of the experimental setup. The frequency comb generator consists of the Ti:sapphire femtosecond laser and the microstructure fiber. SESAM denotes the semiconductor saturable absorber mirror, and PZT piezotransducers. Whereas the repetition rate is phase locked to the hydrogen maser, f_{Ca} and ν_{ceo} are counted. LBO denotes the LiB_3O_5 -frequency doubling crystal, PD photodetectors, PM photomultiplier, PLL phase-locked loop tracking oscillators, and FVC a frequency-to-voltage converter, respectively.

790 nm. The group-velocity dispersion of its cavity is controlled by double-chirped mirrors and an intracavity fused silica prism pair. A similar external prism pair (not shown in Fig. 1) is used for pulse recompression. Approximately 30 mW of the laser output is coupled into a 10-cm-long piece of microstructure fiber with a core diameter of $1.7 \mu\text{m}$ and a zero-GVD wavelength of 780 nm [2]. The output spectrum of the fiber was measured to cover the range from 520 nm to 1100 nm, while the comb structure with distinct lines spaced by 100 MHz was expected to be preserved even in the extreme wings of the SPM-broadened spectrum. This was confirmed by measurement of the fringe visibility (after appropriate optical filtering) at the output of a two-beam interferometer with 100-MHz fringe spacing yielding a 100% visibility throughout the entire comb.

(ii) The repetition frequency of the laser was detected with a fast Si *p-i-n* photo diode yielding a 100-MHz signal with a signal to noise (S/N) ratio of 140 dB ($BW=1$ Hz). It was phase locked to the 100-MHz output signal of a hydrogen maser that was controlled by the PTB primary Cs standards. The actuator of this servo loop is a double piezo device (PZT1A and B) that tilts the output coupler where the spectral components are laterally displaced by the intracavity double-prism GVD compensator. Tilting of the output coupler predominantly modifies the resonator round-trip group delay [9], which, in turn, determines f_{rep} .

(iii) The frequency f_{Ca} of the beat note between the radiation from the Ca standard (at optical frequency ν_{Ca}) and the nearest comb mode was both measured and long-term stabilized. For this purpose, the output of the Ca standard was combined with the emission from the output of the microstructure fiber, which was filtered by an interference filter. The beat note was detected by a Si *p-i-n* photo diode. Its output signal with a typical S/N ratio of 30 dB ($BW=100$ kHz) was filtered by a phase-lock-loop tracking oscillator (PLL) and subsequently counted by a totalizing counter. The beat signal frequency was kept within the hold-in range of

the PLL by an additional servo loop controlling the length of the Ti:sapphire-laser resonator with the help of a piezotransducer (PZT2). The error signal of this slow loop was generated by a frequency-to-voltage converter (FVC) that monitored the output frequency of the PLL. Unwanted cross-talk between the f_{rep} and f_{Ca} servo loops was minimized by adjustment of the lever point of the f_{rep} -tilting actuator to the lateral position of the red comb components at 657 nm. This was accomplished by choosing appropriate gain values (of opposite sign) for the piezotransducers PZT1A and B.

(iv) The frequency ν_{ceo} was measured, as mentioned above, by external self-phase modulation in the microstructure fiber and subsequent second-harmonic generation in a nonlinear optical crystal. The infrared portion of the fiber output around 1070 nm was frequency doubled with a non-critically phase-matched, 10-mm-long LiB_3O_5 (LBO) crystal, which was heated to about 140°C in order to fulfill the type-I phase-matching condition for this wavelength. Possible additional self-phase modulation in the LBO crystal only affects the phase of the ν_{ceo} signal but not the frequency. The beat note between the resulting green SHG signal and the green output of the fiber was detected by a photomultiplier (PM) after spectral and spatial filtering both fields with a single-mode fiber and a 600 l/mm grating, respectively. The PM output signal with a (S/N) ratio of up to 40 dB ($BW=100$ kHz) was filtered with a second tracking oscillator and counted by a second totalizing counter. Both counters were synchronously gated by the same 1-Hz clock signal derived from the 100-MHz hydrogen maser signal.

The optical Ca frequency standard has been described elsewhere [13,15], and only the relevant details shall be given here. This standard is operated in a different building and is linked to the Ti:sapphire laser by a 150-m-long polarization-preserving single-mode fiber. Two independent systems are available, producing clouds of between 10^6 and 10^7 ballistic Ca atoms that are released from magneto-optical traps (MOT 1 and MOT 2). Each trap is loaded for 15 ms.

TABLE I. The differences of the measured Ca $^3P_1-^1S_0$ transition frequencies with respect to the value recommended by CIPM and the absolute and relative uncertainties. Series *A* and *B* refer to the frequency comb generator and previous measurement refers to the harmonic chain.

	$\nu_{\text{Ca}}(\text{expt}) - \nu_{\text{Ca}}(\text{CIPM})$ (Hz)	1σ uncertainty (Hz)	Relative uncertainty
Series <i>A</i>	100	328	7.2×10^{-13}
Series <i>B</i>	148	132	2.9×10^{-13}
Series <i>A + B</i>	126	180	4.0×10^{-13}
Previous measurement [15]	-20	113	2.5×10^{-13}
CIPM recommendation [1]	0	270	6.0×10^{-13}

Then the trapping lasers at 423 nm and the magnetic quadrupole fields are switched off, and a homogeneous magnetic field of 0.23 mT is switched on for spectral separation of the Zeeman components. At the same time the cloud starts to expand and to fall down due to its thermal velocity ($T \cong 3$ mK) and gravity, respectively. After a settling time for the magnetic field of 0.2 ms, the atoms are interrogated by pulses from a frequency-stabilized dye laser. For the frequency measurements MOT 1 was employed, in which the low-velocity atoms are directly captured from a thermal beam by three mutually perpendicular counterpropagating beams with two different frequencies. The atoms were excited in a time-domain analogue of the optical Ramsey excitation by two pulses separated by 216.4 μs followed by two pulses with the same separation from a counterpropagating beam [15]. The frequency of the interrogating laser field was stabilized to the central fringe (full width at half maximum of the fringes 1.16 kHz) of the interference structure that occurs when the fluorescence of the excited atoms is measured as a function of the laser frequency. The uncertainty of the frequency of the Ca optical frequency standard during both measurement series was estimated to 53 Hz. A more detailed uncertainty budget is given in [15]. Several tests have been performed to check the validity of this estimate. By reversing the temporal order of the excitation pulses a shift of a few hertz was determined. The frequency of the laser stabilized to atoms of MOT 1 was compared with its frequency when stabilized to a second MOT 2. Here, the atoms of an effusive beam are decelerated in a Zeeman slower and these slow atoms are deflected and directed towards the magneto-optical trap [16]. The frequency difference measured when the laser was stabilized alternatively to atoms from MOT 1 and MOT 2 by three pulses of a standing wave was 12 ± 15 Hz. An additional laser field at 672 nm was used in MOT 1 to repump the atoms that are lost via the 1D_2 state [17]. The magnetic field of 0.23 mT caused a second-order Zeeman shift of the Ca frequency of +3.1 Hz.

Two measurement series were carried out on different days with a total measurement time of 3000 and 4200 s, respectively. On these days the frequency deviation of the hydrogen maser and the Cs clock resulted in deviations of the measured Ca frequency of +26 Hz and 24 Hz, respectively. The deviations of the corrected measured mean values from the CIPM-recommended value of $\nu_{\text{Ca}}(\text{CIPM}) = 455\,986\,240\,494\,150$ Hz are listed in Table I together with the corresponding relative 1σ uncertainties, which comprise the estimated relative frequency uncertainties of the

standards and those of the frequency measurements. The values of the previous measurement [13] using the harmonic frequency chain and the CIPM values [1] are shown for comparison.

Figure 2 shows the Allan standard deviation of the data of series *B* together with the corresponding frequency instabilities of the Ca standard [18] and those of the hydrogen maser [19]. The inset of Fig. 2 shows a histogram of the data of series *B* (1-s averages) with respect to the CIPM-recommended frequency. The center frequency of the fitted Gaussian of $\nu_{\text{Ca}}(\text{CIPM}) + 46$ Hz agrees within the uncertainty with the mean value of series *B*. The instability of the frequency measurement process is about one order of magnitude larger than the frequency instability of the Ca standard and of the hydrogen maser. So far, the measurements are still limited by the nonoptimum detection of the frequency fluctuations of the Ti:sapphire laser. Substantial improvement of the measurement process can be expected if the instantaneous residual phase error between f_{rep} and the hydrogen maser reference signal is simultaneously measured, i.e., at a high harmonic of f_{rep} .

To conclude, the frequency of an optical frequency standard based on the calcium $^3P_1-^1S_0$ intercombination transition at 657 nm has been measured in terms of the output of a primary cesium frequency standard using an alternative type of optical frequency chain. Accurate frequency measure-

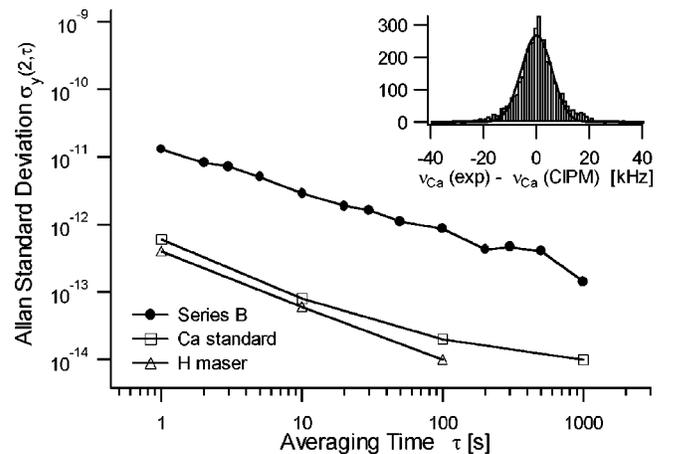


FIG. 2. The Allan standard deviation of measurement series *B* is plotted versus the averaging time τ together with the frequency instabilities of the Ca standard and the hydrogen maser. The inset shows the distribution of the measured values of series *B* (averaging time = 1 s, bin width = 1 kHz, $\sigma = 5.75$ kHz).

ments of this optical standard are of particular importance, since it represents one of the realizations of the unit of length with lowest uncertainty. Furthermore, the optical frequency of the Ca standard is used as a reference for highly accurate frequency measurements of other optical or ultraviolet transitions [20]. The frequency value presented in this Rapid Communication agrees within its relative uncertainty with the CIPM-recommended value, which is based on a measurement with a harmonic frequency chain. The agreement justifies the confidence in the data obtained from both measurement schemes. The advantages of the scheme employed

here, such as the substantially reduced complexity and the accurately known, dense grid of reference frequencies throughout the visible and near-infrared range, allows a variety of new applications including the realization of optical clocks or ultraprecise measurements in fundamental physics.

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