Direct comparison of two cold-atom-based optical frequency standards by using a femtosecond-laser comb

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With a fiber-broadened, femtosecond-laser frequency comb, the 76-THz interval between two laser-cooled optical frequency standards was measured with a statistical uncertainty of 2×10^{-13} in 5 s, to our knowledge the best short-term instability thus far reported for an optical frequency measurement. One standard is based on the calcium intercombination line at 657 nm, and the other, on the mercury ion electric-quadrupole transition at 282 nm. By linking this measurement to the known Ca frequency, we report a new frequency value for the Hg^+ clock transition with an improvement in accuracy of $\sim 10^5$ compared with its best previous measurement.

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State-of-the-art optical frequency standards based on cold atoms and ions exhibit excellent frequency stability and have the potential for achieving high reproducibility and accuracy. Such frequency references should find application in precise tests of fundamental physics and in next-generation atomic clocks. Incorporating these optical-based standards into a clockwork, however, has proved troublesome because their large frequencies (>300 THz) could not be conveniently converted into countable microwave signals. Several optical-frequency measurements have been made with harmonic chains used to multiply the frequency of the 9.2-GHz cesium microwave standard,[1–6] but these chains are complex and their operation requires significant resources. A paradigm-changing simplification was pioneered by Udem *et al.*[7,8] when they used the wide frequency-domain comb output of a femtosecond (fs) mode-locked laser to measure the absolute frequency of the Cs D_1 line and the hydrogen $1s-2s$ transition,[9] the most accurate measurement to date of an optical frequency. A further refinement, by Diddams *et al.*,[10] expanded the available comb spectrum to an optical octave by broadening the fs-laser output in a microstructure fiber, leading to a direct connection between microwave and optical frequencies. A recent comparison of two independent, fiber-broadened fs laser combs that measured the same frequency interval verified that a precision and reproducibility of $<5.1 \times 10^{-16}$ can be attained with this measurement method.[11]

In this Letter we report a high-precision comparison of two promising cold-atom optical frequency standards by use of a fs-laser frequency comb. One standard is based on a 2-mK collection of $\sim 10^7$ neutral ^{40}Ca atoms, and the other probes a single $^{199}Hg^+$ ion that is laser cooled to near the Doppler limit. At 657 nm, a cw frequency-stabilized diode laser is locked to the central Ramsey–Bordé fringe obtained by four-pulse excitation of the Ca $^1S_0-^3P_1$ intercombination transition ($\nu_{Ca} = 456$ THz, $\Delta\nu = 400$ Hz).[12] This system has demonstrated a frequency instability of $4 \times 10^{-15}\ \tau^{-1/2}$ (τ is the averaging time) when it is probing subkilohertz fringe linewidths.[13] For the present measurements the Ca spectrometer was operated with 2.9-kHz linewidths, which gave an estimated short-term instability of $<2 \times 10^{-14}\ \tau^{-1/2}$. The oscillator in the Hg^+ standard is a frequency-narrowed cw dye laser at 563 nm that has a linewidth of $\sim$0.16 Hz for a 20-s integration time.[14] This light is frequency doubled to 282 nm to interrogate the $^2S_{1/2}-^2D_{5/2}$ electric-quadrupole transition ($\nu_{Hg^+} = 1065$ THz, $\Delta\nu = 1.7$ Hz) of a Hg^+ ion that is confined in a linear, cryogenic, rf ion trap. Rabi linewidths as narrow as 6.7 Hz at 282 nm have been observed with this system.[15] For these measurements the laser was stabilized to the Hg^+ ion with a linewidth of 40 Hz at 282 nm, and the instability under these conditions is estimated to be $<3 \times 10^{-15}\ \tau^{-1/2}$.

The fs-comb frequency measurement system shown in Fig. 1 measures the 76-THz interval between the 657- and the 563-nm light which is transported from each stabilized laser by a 10- and a 130-m optical fiber, respectively. No attempt has been made to actively cancel the fiber-added noise,[14,16] which we measured to average as $<4.4 \times 10^{-14}\ \tau^{-1/2}$ for these optical frequencies. The frequency comb is produced by a Kerr-lens mode-locked Ti:sapphire laser that has a bandwidth of 42 nm (FWHM) centered at 810 nm and operates with a repetition rate of $\approx$98 MHz. To control the frequency spacing between comb modes we detect the ninth harmonic of the repetition rate with a signal-to-noise ratio (S/N) of >70 dB in a 100-kHz

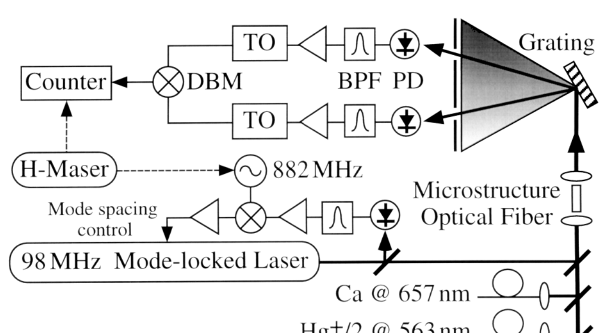

Fig. 1. Block diagram of frequency measurement: DBM, doubly balanced mixer; PD, photodiode; BPF, bandpass filter; TO's, tracking oscillators.

bandwidth (BW) and then mix it with an 882-MHz reference signal from a frequency synthesizer. We phase lock the repetition rate of the mode-locked laser to this reference frequency by using the phase-dependent mixer output to control the horizontal tilt of the high-reflector mirror, which is situated after a dispersion-compensating prism pair.[7–9] The internal clock of the synthesizer is phase locked to a hydrogen maser [H-Maser; $\sigma_y(\tau) \approx 2 \times 10^{-13}\tau^{-1/2}$], whose frequency is known with an uncertainty of $\pm 4 \times 10^{-15}$ by comparison with a Cs primary standard.

To extend the comb bandwidth we launch the fs-laser pulses into a 5 cm-long microstructure optical fiber that has a core diameter of $\approx$1.7 μm and zero group-velocity dispersion near 770 nm.[17] Self-phase modulation and other nonlinearities in the fiber produce an output spectrum from approximately 500 to 1100 nm. The cw light from both stabilized lasers is also coupled into the microstructure fiber to ensure good spatial mode overlap with the fs-laser light. The fiber output is dispersed by an optical grating and imaged onto slits such that only a few modes in the vicinity of the cw light frequencies are selected. A photodiode after each slit detects the rf heterodyne beat note (δ_1, δ_2) between the cw light and a comb mode with a S/N of ~25 dB in a 100-kHz BW, with the background limited by the shot noise of the cw light power. For accurate frequency counting, a tracking oscillator is phase locked to each beat note to provide regenerated signals with >50-dB S/N in a 100-kHz BW. These tracking oscillators consist of a low-phase-noise, voltage-controlled oscillator that is phase locked with an ~100-kHz BW to track the incoming signal in such a way that the broadband, background noise pedestal below the beat note is not reproduced.

In these measurements the comb-mode spacing ν_{rep} is locked tightly to the H-maser frequency, but the offset frequency of the fs-laser comb remains uncontrolled. So, although the comb modes are spaced equidistantly, knowledge of the absolute frequency of an individual comb mode is limited by the frequency jitter of the fs laser (~10 MHz). As we are concerned only with a frequency difference, we can remove this noise that is common to both beat notes by mixing together the correlated signals from the two tracking oscillators.[10] The mixer output is either a stable sum or difference signal $\delta = \delta_1 \pm \delta_2$, which is counted to yield the frequency interval $\Delta\nu = {}^1\!/\!{}_2\,\nu_{\text{Hg}^+} - \nu_{\text{Ca}} = N\nu_{\text{rep}} \pm \delta$. We determined the integer number N and the sign choice for δ unambiguously by comparing $\Delta\nu$ with our previous $\pm$10-MHz measurement of this frequency difference,[18] and we verified our choices by making measurements for different repetition rates.

The inset in Fig. 2 shows a typical time record of the frequency fluctuations of δ, counted with a 5-s gate time. Points that exhibit obvious cycle slip errors, which are due predominantly to a tracking oscillator's losing lock, are eliminated in the data sorting. We calculate the Allan deviation, shown in Fig. 2, for various averaging times by juxtaposing the 5 s gate-time data; the deviation indicates that the measurement precision averages as $(34\ \text{Hz})\tau^{-1/2}$ for the duration of the measurement. Three frequency sources (ν_{Ca}, ν_{Hg^+}, ν_{rep}) contribute to the short-term instability of this measurement, and from these data alone we cannot attribute the noise unambiguously to a specific source. Nonetheless, by assuming that all the noise comes solely from a given source, we can place an upper limit on its short-term Allan deviation. From this datum we infer an upper limit of $\leq 7 \times 10^{-14}\tau^{-1/2}$ for the fractional frequency instability of the two optical standards, although each probably has a significantly better stability.[13–15] Combining the best estimates for the (normalized) instabilities of the microwave and two optical references, as well as for the optical fiber delivery, we arrive at a calculated instability of $3.5 \times 10^{-13}\tau^{-1/2}$ for the 76-THz interval, in good agreement with the measured value of $4.5 \times 10^{-13}\tau^{-1/2}$. There are likely additional degradations of the stability as a result of noise in the microwave detection of the repetition rate and (or) in the synthesizer electronics that multiply the frequency of the H-maser reference.

The results of running the fs-comb measurement system on four separate days over a 6-week period are plotted in Fig. 3 as the frequency offset from the weighted mean, which is $\Delta\nu = 76\,374\,564\,455\,429\,(40)$ Hz. Each of these points represents the weighted mean of the data runs on an individual day, corrected for the second-order Zeeman shifts for both Ca and Hg^+, which are determined to an uncertainty of $<10^{-14}$ for each day. The Ca system contributes to the majority of the 40-Hz uncertainty; the Hg^+ reference supplies a <10-Hz contribution (at 563 nm), limited by the present measurement of the electric-quadrupole shift.[15] The largest systematic error (~30 Hz) stems from uncertainty in our knowledge of the angular overlap of the counterpropagating probe beams in the Ca spectrometer, which leads to a residual first-order Doppler shift when the cold Ca ensemble has a transverse drift velocity.[4] It is noteworthy that the uncertainties for the data of May 5 and May 25 are dominated by systematic effects. The run-to-run measurements on those days are consistent with $\approx$10 Hz, as illustrated for the May 25 data in the Fig. 3 inset, which show only the statistical uncertainty for each run. Given the relatively small

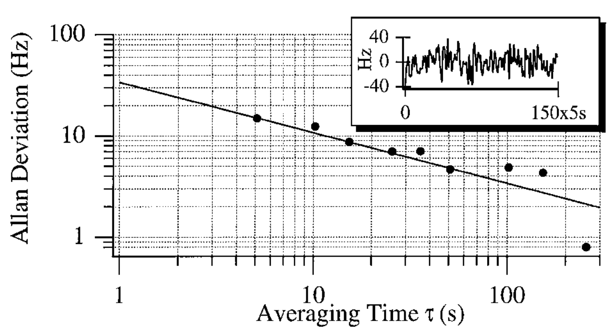

Fig. 2. Allan deviation of a typical measurement record. Curve, $(34\ \text{Hz})\tau^{-1/2}$; inset, the corresponding time record. From these data we place upper limits of $\sigma_y(\tau) \leq 45, 7.4, 6.4 \times 10^{-14}\ \tau^{-1/2}$ for the short-term instability of the microwave reference, ν_{Ca}, and ν_{Hg^+}, respectively.

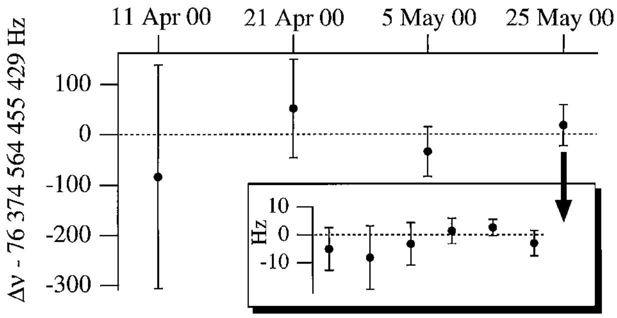

Fig. 3. Frequency deviation (Hz) of $\Delta\nu = {}^{1}\!/_{2}\,\nu_{\mathrm{Hg}^+} - \nu_{\mathrm{Ca}}$. Each data point is the weighted mean of an individual day's data runs. Inset, data runs for May 25 collected over a 50-min period, with each run averaging 230 s. Error bars in the inset reflect only statistical uncertainties.

statistical contribution (~2 Hz on a given day) to the total uncertainty, we believe that fs-laser-based frequency metrology gives us, for the first time, a practical tool for evaluating systematic shifts at an inaccuracy approaching 10^{-15} for these high-accuracy optical standards.

The uncertainties given for the two optical references were estimated for conditions during these experiments only, and no serious attempt was made to minimize systematic effects. Nevertheless, it is encouraging that our results for the frequency difference have a standard deviation of the mean of 60 Hz over a 6-week period. This consistency shows that all three components, the frequency-measurement system and the two optical standards, are reproducible at this level. Thus we offer an improved value for the Hg^+ clock transition frequency by summing our measured $Hg^+/2-Ca$ interval with the absolute frequency of the Ca 657-nm clock transition measured in Ref. 4. We obtain $\nu_{\mathrm{Hg}^+} = 2 \times 532\,360\,804\,949\,559(124)$ Hz, where the uncertainty is dominated by the 120-Hz uncertainty in the Ca measurement. This is an 80,000-fold improvement over the best previous frequency measurement of the 282-nm clock transition.[18]

Work is currently under way to self-reference the frequency offset of our fs comb,[19] and we anticipate confirmation of the Hg^+ and Ca frequencies with a direct rf-to-optical measurement. In fact, locking a mode of the self-referenced comb to one of the optical standards can achieve an all-optical connection between Hg^+ and Ca that eliminates any dependence on the H-maser microwave reference. This interval measurement should then average at the stability of the optical standards alone, allowing for a more precise determination of systematic shifts. The capability to intercompare three high-performance frequency standards (Hg^+, Ca, and Cs) has powerful advantages, such as permitting absolute frequency stabilities to be determined and the fidelity of fs-comb measurements to be tested. Indeed, an optical clock is realized by the repetition-rate output of an all-optically referenced comb, and microwave sources with frequency instabilities near $10^{-15}\,\tau^{-1/2}$ should be obtainable, provided that the repetition-rate signal can be extracted with a suitably high S/N.

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