Femtosecond-laser-based optical clockwork with instability $\leq 6.3 \times 10^{-16}$ in 1 s

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The introduction of femtosecond laser technology into the field of precision optical-frequency metrology has rapidly culminated in a single-step phase-coherent connection between emerging optical frequency standards and the cesium microwave standard on which the SI second is based. By use of femtosecond lasers in combination with microstructure optical fibers, it is now possible to produce an octave-spanning spectrum in the visible and near infrared. Because it originates from a mode-locked laser, this octave-spanning spectrum consists of discrete modes, each of which has a frequency $f_{n} = n f_{r} + f_{0}$, where $f_{r}$ is the pulse repetition frequency, $n$ is an integer, and $f_{0}$ is an offset common to all modes. The frequencies $f_{r}$ and $f_{0}$ can be phase locked to a microwave standard, and the octave-spanning comb operates as a clockwork that permits a single-step, phase-coherent multiplication of the microwave standard up to the optical domain. To date, all tests of the stability and accuracy of the clockwork have been performed with a microwave frequency reference.

Alternatively, the octave-spanning comb can be phase locked to an optical-frequency standard, thereby operating as a clockwork to transfer the stability and accuracy of an optical standard down to a countable microwave frequency. In this case, the precision of the clockwork must not degrade the exceptionally low fractional frequency instability of the optical standard, which can be $\leq 4 \times 10^{-15}$ in 1 s of averaging. Diddams et al. recently demonstrated an optical atomic clock based on this concept, with instability of $\leq 7 \times 10^{-15}$ in 1 s. However, in that case the measure of instability was limited by the present configuration of the Ca standard that was employed, so potential limitations due to the clockwork remained untested. In this Letter, we report testing the femtosecond-laser-based clockwork directly and establishing an upper limit of $6.3 \times 10^{-16}$ for its 1-s instability. We further verify that the frequencies of the comb elements across an entire octave are equal to their expected phase-locked values, with an uncertainty of $4 \times 10^{-17}$. This upper limit represents an improvement of more than a factor of 10 over the best previous test of the uncertainty of the nonlinearly generated comb frequencies of which we are aware. Furthermore, the results presented here conclusively demonstrate that the stability and accuracy of the highest-quality optical standards can be faithfully transferred to hundreds of thousands of individual comb elements across the entire visible and near-infrared spectrum. This transferability is critical to the development of optical clocks because the frequency spacing ($f_{r}$) between the modes is the clock's countable output, which should then possess the same stability and accuracy as the optical standard.

The concept of our measurement is to phase-coherently lock all elements of two octave-spanning frequency combs to a stable cw reference laser (the two combs have the same value of $f_{r}$ but different values of $f_{0}$). Subsequently, we measure and analyze the heterodyne beats between the two combs in different spectral regions to determine how precisely they track the reference laser (Fig. 1). Since the noise of the cw laser is common to both combs, we can evaluate the precision of the various phase-locked loops as well as the stability and frequency accuracy of the comb elements that are nonlinearly generated in the microstructure optical fiber. In comparing the combs...
at optical frequencies as opposed to the frequency of \( f_r \), we gain a factor of nearly \( 10^6 \) (the mode number) in measurement resolution.

More-thorough descriptions of the generation and control of the frequency comb can be found in Refs. 8 and 11. Here we present only the most-relevant details. Each frequency comb is generated by means of coupling 250–300 mW of the output of a mode-locked Ti:sapphire laser\(^{12} \) (\( f_\text{r} = 750 \text{ MHz} \)) into microstructure optical fibers\(^{13} \) that are 20–30 cm in length. Although they are similar in concept, the independent femtosecond comb generators differ in critical features. For example, the two systems have different laser and servo construction and employ different lengths of microstructure fibers with different coupled peak powers.

The spectrum out of each fiber spans the octave from ~500 to 1100 nm. We use the self-referencing technique\(^{6} \) to determine \( f_\text{fb} \) for each comb by frequency doubling the infrared components and heterodyning them with the visible components. We then phase lock \( f_\text{fb} \) for each system to a stable radio frequency synthesized from a hydrogen maser (instability, \( \sim 2 \times 10^{-15} \) in 1 s), using the pump laser’s power as the actuator.\(^{5} \) This phase-locked optical beat is monitored with a high-resolution counter, and the typical standard deviation in 1 s for each system is \( \approx 25 \text{ mHz} \) (\( \approx 5 \times 10^{-17} \)). With \( f_\text{fb} \) fixed in this manner, we control the other degree of freedom (\( f_\text{r} \)) of each comb by measuring and phase locking the heterodyne beat (\( f_\text{fb} \)) between one element of the comb at 456 THz and a cavity-stabilized diode laser. In this case, a piezo-mounted mirror is used as the actuator. Because the comb elements from the femtosecond laser already have a well-defined phase relationship, phase locking one mode to the diode laser in principle locks all the modes to the diode laser. Indeed, the data presented here verify this concept to a high degree of precision. Again, we count the phase-locked optical beat for each system and find a typical standard deviation of \( \approx 100 \text{ mHz} \) in 1 s. These data alone imply that every mode of the octave-spanning combs tracks the diode laser with a relative uncertainty of \( \approx 2 \times 10^{-16} \). We have made no attempt to orthogonalize the control of \( f_\text{fb} \) and \( f_\text{r} \) to reach this level, although doing so might improve the performance.\(^{14} \)

Counting of the phase-locked beats as just described directly verifies the stability of the comb element at only the 456-THz frequency of the diode laser. A much more rigorous test involves the comparison of the two combs across their octave spans. To accomplish this, we offset the combs by an amount \( \Delta f_\text{fo} = f_{\text{o1}} - f_{\text{o2}} = 120 \text{ MHz} \) in the phase-locked loops, while leaving \( f_\text{r} \) the same for both combs. We then spatially and temporally overlap portions of the beams from each system on a photodiode to measure the heterodyne beat between the two combs. We set the temporal overlap by adjusting the phase-locked value of \( f_\text{fb} \) in one system such that \( f_\text{r} \) differs slightly from that of the second system. With a fast oscilloscope, we monitor the arrival times of the two pulse trains as they come into coincidence on the photodiode. When exact coincidence is achieved, a strong beat at \( \Delta f_\text{fo} = 120 \text{ MHz} \) (signal/noise ratio, \( \simeq 40 \text{ dB} \) in 300-kHz bandwidth) is observed and the phase-locked value of \( f_\text{fb} \) is reset so that \( f_\text{r} \) is again equal in the two systems. This beat is bandpass filtered (6-MHz bandwidth), amplified, and counted.

Using optical filters in conjunction with Si, GaAs, and InGaAs detectors, we have measured the absolute value of \( \Delta f_\text{fo} \) and its instability at 550, 350, and 275 THz. The results are summarized in Table 1. The Allan deviation (a measure of fractional frequency instability) computed from the counter readings of \( \Delta f_\text{fo} \) at 550 THz is shown in Fig. 2. When we compute the Allan deviation from data acquired with counter gate times of 1, 3, and 10 s we see a dependence of close to \( \tau^{-1} \), which is expected for white phase noise with a rms phase fluctuation that is constant in time. We

Table 1. Summary of Measured Stability and Offset Between the Two Combs Locked to a Common Laser Diode at 456 THHz

<table>
<thead>
<tr>
<th>Frequency (THz)</th>
<th>1-s Allan Deviation (×10^{-16})</th>
<th>Fractional Offset (×10^{-17})</th>
<th>Integration Time (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>275</td>
<td>7.0</td>
<td>(-0.5 ± 2.9)</td>
<td>539</td>
</tr>
<tr>
<td>350</td>
<td>7.2</td>
<td>(5.6 ± 3.0)</td>
<td>500</td>
</tr>
<tr>
<td>550</td>
<td>8.9</td>
<td>(-2.0 ± 1.5)</td>
<td>1186</td>
</tr>
</tbody>
</table>

*aThe fractional offset values at each frequency are the weighted averages of the respective data of Fig. 3. All data were acquired during a period of \( \sim 10 \text{ h} \).*

![Fig. 2. Allan deviation of \( \Delta f_\text{fo} \) at 550 THz. The squares are the Allan deviation computed from the time series of 1-s counter readings. The triangles are the Allan deviation computed by means of changing the counter’s gate time.](image-url)
then established an upper limit of $6.3 \times 10^{-16}$ for the 1-s instability of the femtosecond-laser-based clockwork. However, it is very likely that this upper limit arises not from the comb but from the uncontrolled fluctuations in path length between the two laser systems. For example, fluctuations of a few hundred nanometers on a 1-s time scale as a result of vibrating mechanics or air currents would lead to a fractional instability of $\sim 7 \times 10^{-16}$.

The offset of the measured value of $\Delta f_0$ from the expected 120 MHz provides information about possible frequency errors that might occur in the nonlinear generation of the octave-spanning comb. This is a particularly important point, since this clockwork ultimately needs to be capable of supporting future optical standards with projected fractional frequency uncertainties approaching $1 \times 10^{-18}$. The best previous test of the actual frequencies of the elements of an octave-spanning comb demonstrated an upper-limit uncertainty of $5 \times 10^{-16}$. Tests of the uniformity of femtosecond-laser-based frequency combs have shown remarkable uncertainties as low as $3 \times 10^{-18}$, however, those tests did not control $f_0$ and employed spectra broadened to only 44 THz in standard silica fiber. At each optical frequency we made four measurement sets of the offset of $\Delta f_0$ from 120 MHz (Fig. 3). The error bars on each point indicate the gate time and the number of counter readings in the measurement set. The largest offset is found at 350 THz, at which the average offset is $19 \pm 11$ mHz, or fractionally $(5.6 \pm 3.0) \times 10^{-17}$. A shift of this order could be due to the thermal expansion of the optical table as a result of a temperature change of 1°C in the laboratory over 1 h. The weighted average of the three values in Table 1 provides an offset from 120 MHz of 0.14 mHz, with an uncertainty of $\pm 5$ mHz [fractionally $(0.04 \pm 1.3) \times 10^{-17}$ at 400 THz]. Since we cannot yet verify the source of the offset at 350 THz, we adopt the scatter of the offsets given in Table 1 as the average uncertainty in the frequencies of the comb lines, which is $4 \times 10^{-17}$ at 400 THz.

In conclusion, we have demonstrated that the stability and accuracy limits of an octave-spanning comb generated with a femtosecond laser are at a sufficiently low level to be useful as a clockwork for the best current optical-frequency standards. As the current results are at the limit imposed by Doppler shifts, active control of all optical paths will be necessary in the future to reach the ultimate stability and accuracy limits.

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References