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

Scott A. Diddams and L. Hollberg

National Institute of Standards and Technology, 325 Broadway, Boulder, Colorado 80305

Long-Sheng Ma

JILA, University of Colorado, Boulder, Colorado 80309

Lennart Robertsson

Bureau International des Poids et Mesures, Pavillon de Breteuil, 92312 Sevres, France

Received July 6, 2001

Two octave-spanning optical-frequency combs (750-MHz comb spacing) are phase locked to a common continuous-wave laser diode. The measured instability of the heterodyne beat between the two combs demonstrates that the intrinsic fractional frequency noise of a comb is $\leq 6.3 \times 10^{-16}$ in 1 s of averaging across the ~ 300 -THz bandwidth. Furthermore, the average frequencies of the elements of the two combs are found to agree within an uncertainty of 4×10^{-17} across the entire octave. We demonstrate the possibility of transfering the stability and accuracy of the best current optical standards to $\sim 500,000$ individual oscillators across the visible and near-infrared spectrum.

OCIS codes: 320.7090, 120.3940, 140.4050.

The introduction of femtosecond laser technology into the field of precision optical-frequency metrology^{1,2} 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.^{3,4} 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 frequency $f_n = nf_r + f_o$, where f_r is the pulse repetition frequency, n is an integer, and f_o is an offset common to all modes. The frequencies f_r and f_o 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.5-8

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.^{9,10} Diddams *et al.* recently demonstrated an optical atomic clock based on this concept, with instability of $\leq 7 \times 10^{-15}$ in 1 s.¹¹ 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 6.3×10^{-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 4×10^{-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 frequencies⁵ 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 nearinfrared 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 phasecoherently 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_o). 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



Fig. 1. Comparison of two octave-spanning combs that are phase locked to a common laser diode. BP, bandpass; Spec., spectrum.

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 laser¹² ($f_r = 750$ MHz) into microstructure optical fibers¹³ 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 \sim 500 to 1100 nm. We use the self-referencing technique⁴ to determine f_o for each comb by frequency doubling the infrared components and heterodyning them with the visible components. We then phase lock f_o for each system to a stable radio frequency synthesized from a hydrogen maser (instability, $\sim 2 \times 10^{-13}$ in 1 s), using the pump laser's power as the actuator.⁵ This phase-locked optical beat is monitored with a highresolution counter, and the typical standard deviation in 1 s for each system is $\leq 25 \text{ mHz} (\leq 5 \times 10^{-17})$. With f_o fixed in this manner, we control the other degree of freedom (f_r) of each comb by measuring and phase locking the heterodyne beat (f_b) 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 phase 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 $\leq 100 \text{ mHz in 1 s.}$ These data alone imply that every mode of the octave-spanning combs tracks the diode laser with a relative uncertainty of $\leq 2 \times 10^{-16}$. We have made no attempt to orthogonalize the control of f_o and f_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 $\Delta f_o = f_{o1} - f_{o2} =$ 120 MHz in the phase-locked loops, while leaving f_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_b in one system such that f_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 $\Delta f_o = 120$ MHz (signal/noise ratio, ≥40 dB in 300-kHz bandwidth) is observed and the phase-locked value of f_b is reset so that f_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 Δf_o 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 Δf_o 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 τ^{-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 CombsLocked to a Common Laser Diode at 456 THz^a

Frequency (THz)	$\begin{array}{c} \text{1-s Allan} \\ \text{Deviation} \\ (\times 10^{-16}) \end{array}$	$\begin{array}{c} \textbf{Fractional} \\ \textbf{Offset} \\ (\times 10^{-17}) \end{array}$	Integration Time (s)
$\begin{array}{c} 275\\ 350\\ \overline{}\end{array}$	7.0 7.2	$(-0.5 \pm 2.9) \ (5.6 \pm 3.0)$	539 500
550	8.9	(-2.0 ± 1.5)	1186

^{*a*}The 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 ~ 10 h.



Fig. 2. Allan deviation of Δf_o 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.



Fig. 3. Measured offset of the two combs from the 120-MHz phase-locked value for three different spectral regions. The weighted average of all data is 0.14 ± 5 mHz at the optical frequencies.

note that the incorrect $\tau^{-1/2}$ dependence is measured when the Allan deviation for $\tau > 1$ s is computed from the juxtaposition of a long series of 1-s samples.¹⁵ We further find that the 1-s Allan deviation differs by no more than 30% for the three measurement points across the octave. Since both combs contribute to the Allan deviation of Δf_o , we can assume that the fluctuations in a single comb are less by a factor of $\sqrt{2}$. Taking the Allan deviation at 550 THz, we then established an upper limit of $6.3 imes10^{-16}$ 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 Δf_o 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 1×10^{-18} .¹⁶ The best previous test⁵ of the actual frequencies of the elements of an octave-spanning comb demonstrated an upper-limit uncertainty of 5×10^{-16} . Tests of the uniformity of femtosecond-laser-based frequency combs have shown remarkable uncertainties as low as 3×10^{-18} ; however, those tests did not control f_o and employed spectra broadened to only 44 THz in standard silica fiber.¹⁷ At each optical frequency we made four measurement sets of the offset of Δf_o 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 ± 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 ± 5 mHz [fractionally (0.04 \pm 1.3) \times 10¹⁷ 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 4×10^{-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.

The authors acknowledge the thoughtful comments and assistance of Th. Udem, J. Bergquist, J. Ye, J. L. Hall, C. Oates, and A. Curtis. We are indebted to R. S. Windeler of Lucent Technologies for providing the microstructure fiber and are grateful to A. Bartels of GigaOptics GmbH for his assistance with the femtosecond laser. S. A. Diddams's e-mail address is sdiddams@boulder.nist.gov.

References

- 1. Th. Udem, J. Reichert, R. Holzwarth, and T. W. Hänsch, Phys. Rev. Lett. 82, 3568 (1999).
- Th. Udem, J. Reichert, R. Holzwarth, and T. W. Hänsch, Opt. Lett. 24, 881 (1999).
- S. A. Diddams, D. J. Jones, J. Ye, S. T. Cundiff, J. L. Hall, J. K. Ranka, R. S. Windeler, R. Holzwarth, Th. Udem, and T. W. Hänsch, Phys. Rev. Lett. 84, 5102 (2000).
- D. J. Jones, S. A. Diddams, J. K. Ranka, R. S. Windeler, A. J. Stentz, J. L. Hall, and S. T. Cundiff, Science 228, 635 (2000).
- R. Holzwarth, Th. Udem, T. W. Hänsch, J. C. Knight, W. J. Wadsworth, and P. St. J. Russell, Phys. Rev. Lett. 85, 2264 (2000).
- Th. Udem, S. A. Diddams, K. R. Vogel, C. W. Oates, E. A. Curtis, W. D. Lee, W. M. Itano, R. E. Drullinger, J. C. Bergquist, and L. Hollberg, Phys. Rev. Lett. 86, 4996 (2001).
- J. Stenger, Ch. Tamm, N. Haverkamp, S. Weyers, and H. R. Telle, http://xxx.lanl.gov/abs/physics/0103040.
- S. A. Diddams, Th. Udem, K. R. Vogel, C. W. Oates, E. A. Curtis, R. S. Windeler, A. Bartels, J. C. Bergquist, and L. Hollberg, Proc. SPIE 4269, 77-83 (2001).
- C. W. Oates, E. A. Curtis, and L. Hollberg, Opt. Lett. 25, 1603 (2000).
- B. C. Young, F. C. Cruz, W. M. Itano, and J. C. Bergquist, Phys. Rev. Lett. 82, 3799 (1999).
- S. A. Diddams, Th. Udem, J. C. Bergquist, E. A. Curtis, R. E. Drullinger, L. Hollberg, W. M. Itano, W. D. Lee, C. W. Oates, K. R. Vogel, and D. J. Wineland, Science 293, 825 (2001).
- 12. A. Bartels, T. Dekorsy, and H. Kurz, Opt. Lett. 24, 996 (1999).
- J. K. Ranka, R. S. Windeler, and A. J. Stentz, Opt. Lett. 25, 25 (2000).
- J. Ye, L.-S. Ma, and J. L. Hall, "Molecular iodine clock," Phys. Rev. Lett. (to be published).
- 15. P. Lesage, IEEE Trans. Instrum. Meas. 32, 204 (1983).
- A. A. Madej and J. E. Bernard, in *Frequency Measurement and Control*, A. N. Luiten, ed. (Springer-Verlag, Berlin, 2001), pp. 153–194.
- R. Holzwarth, "Measuring the frequency of light using femtosecond laser pulses," Ph.D. dissertation (Ludwig-Maximilians-Universität, Munich, Germany, 2001).