Frequency Uncertainty for Optically Referenced Femtosecond Laser Frequency Combs

Long-Sheng Ma, Zhiyi Bi, Albrecht Bartels, Kyoungsik Kim, Lennart Robertsson, Massimo Zucco, Robert S. Windeler, Guido Wilpers, Chris Oates, Leo Hollberg, and Scott A. Diddams, *Member, IEEE*

***Abstract*—We present measurements and analysis of the currently known relative frequency uncertainty of femtosecond laser frequency combs (FLFCs) based on Kerr-lens mode-locked Ti:sapphire lasers. Broadband frequency combs generated directly from the laser oscillator, as well as octave-spanning combs generated with nonlinear optical fiber are compared. The relative frequency uncertainty introduced by an optically referenced FLFC is measured for both its optical and microwave outputs. We find that the relative frequency uncertainty of the optical and microwave outputs of the FLFC can be as low as 8×10^{-20} and 1.7×10^{-18}, with a confidence level of 95%, respectively. Photo-detection of the optical pulse train introduces a small amount of excess noise, which degrades the stability and subsequent relative frequency uncertainty limit of the microwave output to 2.6×10^{-17}.**

***Index Terms*—Femtosecond laser, frequency comb, optical frequency metrology.**

I. INTRODUCTION

A FEMTOSECOND laser frequency comb (FLFC) consists of the broadband array of optical frequencies generated by a mode-locked femtosecond laser. When referenced to an optical or microwave frequency standard, the FLFC can operate as an extremely broadband phase-coherent frequency synthesizer with three principle functions: optical-to-optical frequency synthesis, optical-to-microwave frequency synthesis, and microwave-to-optical frequency synthesis [1]. In this role, the FLFC technique has found wide-spread use in optical frequency metrology [2], emerging optical atomic clocks [3], [4], new techniques of spectroscopy [5], [6], and low noise time [7], [8] and frequency domain waveform synthesis [9], [10]. The FLFC plays an increasingly important role in the comparison of laboratory-based frequency standards, enabling the measurement of fundamental physical constants in addition to searches for possible time-variations of the same [11]–[15]. Moreover, the frequency-domain control of the FLFC provides access to the evolution of the carrier-envelope phase in the time domain [16], which has been the key to the recent experiments on high-field interactions that depend on the exact phase between the carrier and the envelope of few-cycle laser pulses [17], [18].

Within the context of these various applications, it is important to investigate the potential limitations of different types of FLFCs. In this paper we are primarily interested in exploring the frequency uncertainty of the FLFC when referenced to an optical frequency standard—an approach that provides the highest performance. Recent experiments have shown that with suitable servo-control, the short-term (<1 s) frequency noise level of the FLFC can be significantly decreased, allowing the generation of FLFC optical modes with linewidths at the hertz-level[19] and microwave signals with very low phase noise close to the carrier [10]. In other work, the long-term (typical averaging times >1000 s) frequency stability of the FLFC has been addressed. A few such tests have been performed using microwave standards to reference FLFCs, which were subsequently compared in the optical domain (i.e., microwave-to-optical synthesis) [20]–[22], resulting in relative uncertainty as low as 5×10^{-16}. Referencing the FLFC to an optical standard provides improved stability allowing shorter averaging time, leading to lower uncertainty. In a recent report [23], we demonstrated that in such a configuration the relative frequency uncertainty in the output comb modes of the FLFC is near 1×10^{-19}. The reproducibility of this performance was verified by comparison of four combs of different construction from three laboratories. Results at this level have also been obtained with the unstabilized FLFC used as a "transfer oscillator." In this scheme, a judicious choice of frequency mixings effectively eliminates the noise of the femtosecond laser when it is used to determine the ratio of widely separated optical frequencies [24], [25].

Here, we expand on our earlier results [23], providing additional data that further reduce the relative uncertainty of the FLFC to 8×10^{-20} when used for optical-to-optical synthesis.

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L.-S. Ma is with Bureau International des Poids et Mesures, Pavillon de Breteuil, 92312 Sevres, France, and also with the Key Laboratory of Optical and Magnetic Resonance Spectroscopy, East China Normal University, Shanghai 200062, China (e-mail: lsma@ecnu.edu.cn).

Z. Bi is with Key Laboratory of Optical and Magnetic Resonance Spectroscopy, East China Normal University, Shanghai 200062, China.

A. Bartels was with the National Institute of Standards and Technology, Boulder, CO 8030 USA. He is now with Gigaoptics GmbH, Konstanz 78462, Germany.

K. Kim was with the National Institute of Standards and Technology, Boulder, CO 8030 USA. He is now with the School of Mechanical Engineering, Yonsei University, Seoul 120-749, Korea.

L. Robertsson is with Bureau International des Poids et Mesures, Pavillon de Breteuil, 92312 Sevres, France.

M. Zucco was with Bureau International des Poids et Mesures, Pavillon de Breteuil, 92312 Sevres, France. He is now with the National Institute of Meterological Research, Turin 10135, Italy.

R. S. Windeler is with OFS Laboratories, Murray Hill, NJ 07974 USA.

G. Wilpers was with National Institute of Standards and Technology, Boulder, CO 80305 USA. He is now with National Physical Laboratory, Teddington, Middlesex TW11 0LW, U.K. (e-mail: sdiddams@boulder.nist.gov).

C. Oates, L. Hollberg, and S. A. Diddams are with National Institute of Standards and Technology, Boulder, CO 80305 USA (e-mail: sdiddams@boulder.nist.gov).

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We also include new results of our measurements to determine the relative uncertainty in the optical-to-microwave conversion performed with the FLFC. In this case, data are provided for both the optical pulse train produced by the FLFC and the subsequent microwave signal obtained after photo-detection of the optical pulse train.

II. EXPERIMENTAL SETUP

The data presented here were collected from four FLFCs constructed at the three different institutes participating in these measurements. FLFCs of the type shown in Fig. 1(a) were constructed at the Bureau International de Poids et Mesures (BIPM) and the East China Normal University (ECNU). These FLFCs are transportable devices and were brought to the National Institute of Standards and Technology (NIST) for measurements that took place in 2003. These two FLFCs are referred to as BIPM-C2 and ECNU-C1, respectively. At NIST, two FLFCs of the type shown in Fig. 1(b) were employed in these measurements. The NIST FLFCs are referred to as NIST-BB1 and NIST-BB2. In all cases, two phase-lock-loops are employed to servo-control the FLFC carrier-envelope offset frequency (f_{ceo}) and the repetition rate (f_{rep}), which is the frequency spacing of the modes of the FLFC. Details of the different FLFCs and their control are provided below. During the period of August-October in 2003, NIST-BB2, BIPM-C2, and ECNU-C1 were compared with NIST-BB1. Subsequently, the comparisons were continued between NIST-BB2 and NIST-BB1 in April and December of 2004, and April of 2005. For these later measurements, the environmental isolation of the NIST FLFCs was improved by enclosing the lasers and critical beam paths in boxes and tubes.

A. *FLFC Employing Nonlinear Microstructured Fiber*

The two transportable combs (BIPM-C2 and ECNU-C1) constructed in the BIPM and ECNU are based on a six-mirror ring laser [26] as shown in Fig. 1(a). One of the mirrors is mounted on a fast piezo-electric transducer (PZT) that is used to control the repetition rate f_{rep}. A second mirror is mounted on a long PZT, which is used to compensate the long-term drift of the femtosecond laser cavity. Each femtosecond laser is pumped by a solid state laser at 532 nm with a pump power of about 5 W, giving a laser output power of approximately 500 mW. The width of the femtosecond laser output spectra are expanded to cover a full optical octave by a nonlinear microstructured fiber [27] having a length of ~30 cm. The infrared comb light near 1064 nm is frequency doubled to green light in a 5-mm-long $\mathrm{KNbO_3}$ crystal and then mixed on a photodiode with the green comb light near 532 nm. This generates the beat signal for the control of f_{ceo}, having a typical signal-to-noise ratio (SNR) of 35 dB in 300-kHz resolution bandwidth. Feedback to an acoustooptic modulator (AOM) in the path of the pump beam is used to change the laser power, thereby controlling f_{ceo}. The femtosecond laser, nonlinear microstructured fiber, and self referencing set-up are entirely contained in a sealed aluminum box with dimensions $69 \times 54 \times 23$ cm^3. Using various dichroic beamsplitters, the transportable combs were designed to have three output beams each in different regions of the optical spectrum near 532, 600–900, and 1064 nm.

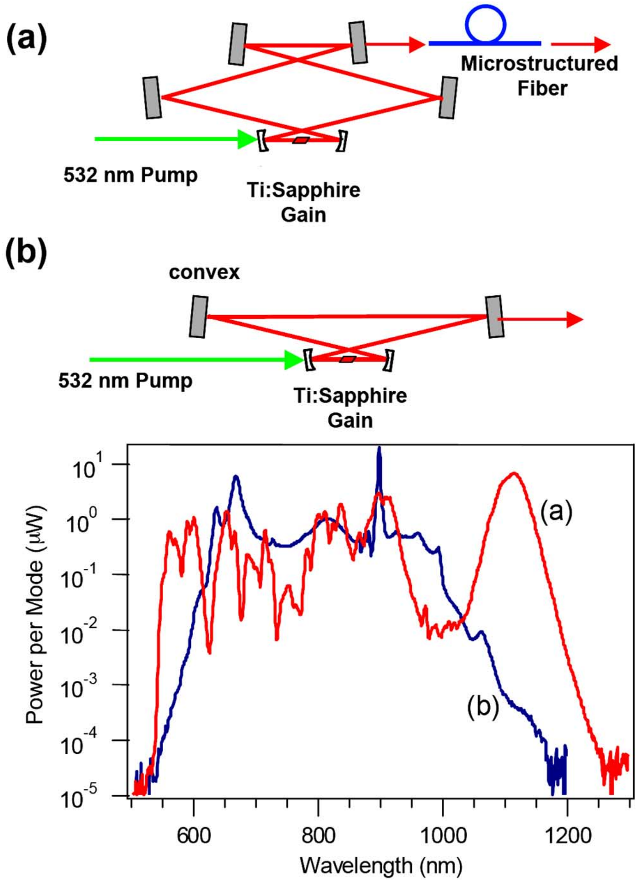

Fig. 1. Diagram of two types of FLFC employed in this work. (a) Femtosecond laser that emits a spectrum that is subsequently expanded in nonlinear microstructured fiber. (b) Femtosecond ring laser that directly emits a broadband continuum. The spectra from these two systems are shown in the lower part of the figure, where the power per 1 GHz mode is plotted as a function of wavelength.

B. *Broadband FLFC*

The NIST FLFCs (NIST-BB1 and NIST-BB2) are based on four-mirror ring lasers [Fig. 1(b)] that directly emit a broadband spectrum spanning the range ~(560–1150) nm at −50 dB below the maximum [28]. The total average output power is ~640 mW with 8 W of pump light at 532 nm. Additional broadening in a nonlinear microstructure fiber is therefore not required. Instead, the f_{ceo} can be measured by frequency tripling light emitted near 960 nm and heterodyning it with frequency doubled light near 640 nm [29]. The two UV beams at 320 nm are coupled into a single mode fiber that provides good mode matching to efficiently generate a beat signal at f_{ceo}, with typical SNR of 25 dB in 300-kHz bandwidth. Similar to the case above, both of these lasers have a piezo-mounted mirror and an AOM in the pump beam as servo actuators. Further details are provided in reference [29]. We note that other broadband frequency combs exist.

C. *Precision Control of the FLFC With an Optical Reference*

The f_{rep} and f_{ceo} must be controlled precisely in the FLFC in order to control the comb modes over the entire spectrum. For microwave-to-optical synthesis, f_{rep} and f_{ceo} can be phase-locked to a microwave frequency standard such as a Cs atomic

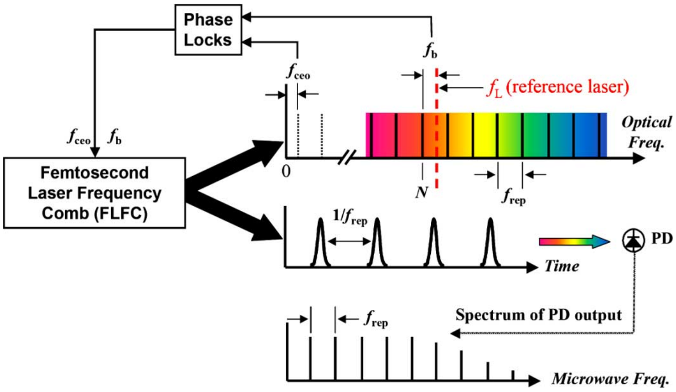

Fig. 2. FLFC in both the time and frequency domains. The output of a femtosecond laser consists of a broad frequency comb. Two phase-locks are employed to control $f_{\rm ceo}$ and $f_{\rm rep}$ relative to an optical reference laser $(f_{\rm L})$. The time-domain output is a repetitive train of optical pulses. When detected with a fast photodiode (PD), the result is a comb of frequencies in the microwave domain extending from dc up to the bandwidth of the PD. In this work, we search for possible deviations of the optical frequency comb, the associated time-domain pulse train, and the microwave frequency comb.

clock or a hydrogen maser. Alternatively, the FLFC can serve as an optical gear to divide down an optical frequency standard to a countable microwave frequency, or to compare various optical frequency standards separated by a gap of up to hundreds of terahertz in the optical domain. In this case, the FLFC is phase-locked to an optical reference as shown in Fig. 2. For our experiments, a cavity-stabilized diode laser at 657 nm [30] was used as a reference laser. The beat signal $f_{\rm b}$ between the reference laser and an adjacent comb line N_L serves as a control signal for the optical phase-lock servo. With $f_{\rm ceo}$ already controlled, this optical phase lock effectively acts to fix the frequencies of the other modes of the FLFC in addition to $f_{\rm rep}$, which can be written as

$$f_{\rm rep} = (f_L - f_{\rm ceo} - f_b)/N_L \tag{1}$$

where $f_{\rm b}$ is the beat frequency between $f_{\rm L}$ and mode N_L of the the FLFC.

To verify the precision of the control of the FLFC, in all measurements we use auxiliary frequency counters to monitor both of the phase-locked beats $f_{\rm ceo}$ and $f_{\rm b}$[31]. Fig. 3 shows the typical stability of these beats for the NIST-BB1 system. The performance was similar for the other systems employed. Both $f_{\rm ceo}$ and $f_{\rm b}$ are radio frequencies (a few hundred megahertz) that add or subract from the optical elements of the FLFC, so by the measure of Fig. 3, the phase locks contribute noise at the millihertz level in 10 s, which averages down to the level of tens of microhertz. On an optical frequency of roughly 500 THz, this corresponds to a relative instability introduced by the phase-locks at or below the level of 1×10^{-19}. While such stability does not imply a similarly small frequency uncertainty in the modes of the FLFC, it is a necessary requirement.

III. MEASUREMENT AND RESULTS

We search for potential limitations in the FLFCs by rigorously comparing the four different systems described above. The basic scheme of our measurements is to compare pairs of femtosecond laser synthesizers (labeled by indexes 1 and 2) and verify with (a) optical heterodyne techniques, (b) nonlinear cross correlation, and (c) photodetection of $f_{\rm rep}$ that the output modes and repetition rate have their expected frequencies relative to the continuous wave reference laser having frequency f_L = 456 THz. In what follows, we will present and discuss the results from these three types of comparisons.

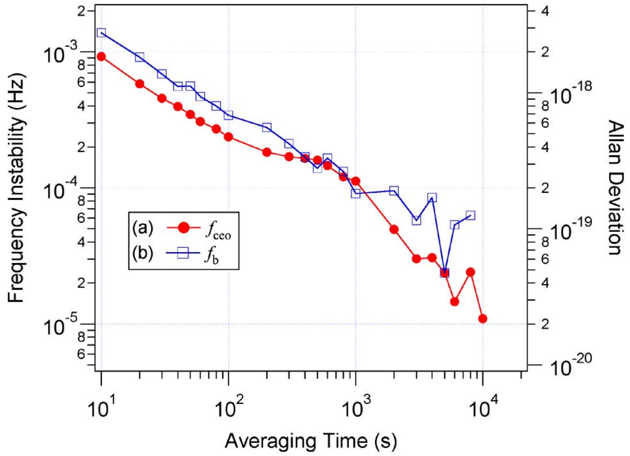

Fig. 3. Tracking capability of a FLFC to a cavity-stabilized reference laser at 657 nm. (a) $f_{\rm ceo}$ control for NIST-BB1. (b) $f_{\rm b}$ for repetition rate control of NIST-BB1. The gate time for the counter is 10 s and the instability at longer averaging times is computed by averaging groups of adjacent 10 samples. Combined with dead time in the counters, this leads to the departure from the expected $1/\tau$ dependence $(\tau = \text{averaging time})$.

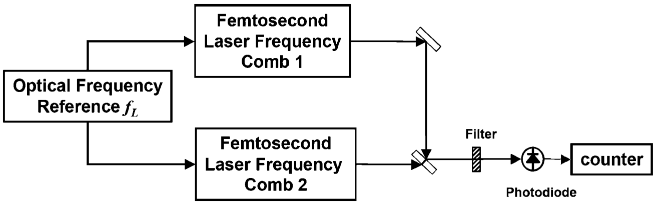

Fig. 4. Comparison of comb line frequencies by optical heterodyne measurements between two optical phase controlled FLFCs.

A. *Optical Heterodyne Comparisons of FLFC Spectral Lines*

Fig. 4 shows the configuration for the optical frequency comparisons of FLFC spectral lines by heterodyne detection. When the FLFC is phase locked on the cavity-stabilized diode laser with frequency f_L , the frequencies of the spectral lines from the two FLFCs are given by

$$f_1(N_1) = f_{\rm ceo1} + N_1 \times f_{\rm rep1} \tag{2}$$

$$f_2(N_2) = f_{\rm ceo2} + N_2 \times f_{\rm rep2} \tag{3}$$

where N_1 and N_2 assume integer values as the mode indexes of the two combs, and $f_{\rm rep1}$ and $f_{\rm rep2}$ are given by (1).

In these experiments, we compare modes of the comb that share the same index, thereby requiring $N_1 = N_2 = N$ (integer values). Thus, the difference of the repetition rates $\Delta f_{\rm rep}$ and the optical frequencies $\Delta f = f_1 - f_2$ between the two FLFCs are independent of the frequency f_L. The beat frequency between the comb lines of two FLFCs can be given by

$$\Delta f = f_1(N_1) - f_2(N_2) = (f_{\rm ceo1} - f_{\rm ceo2}) + N \times \Delta f_{\rm rep}. \tag{4}$$

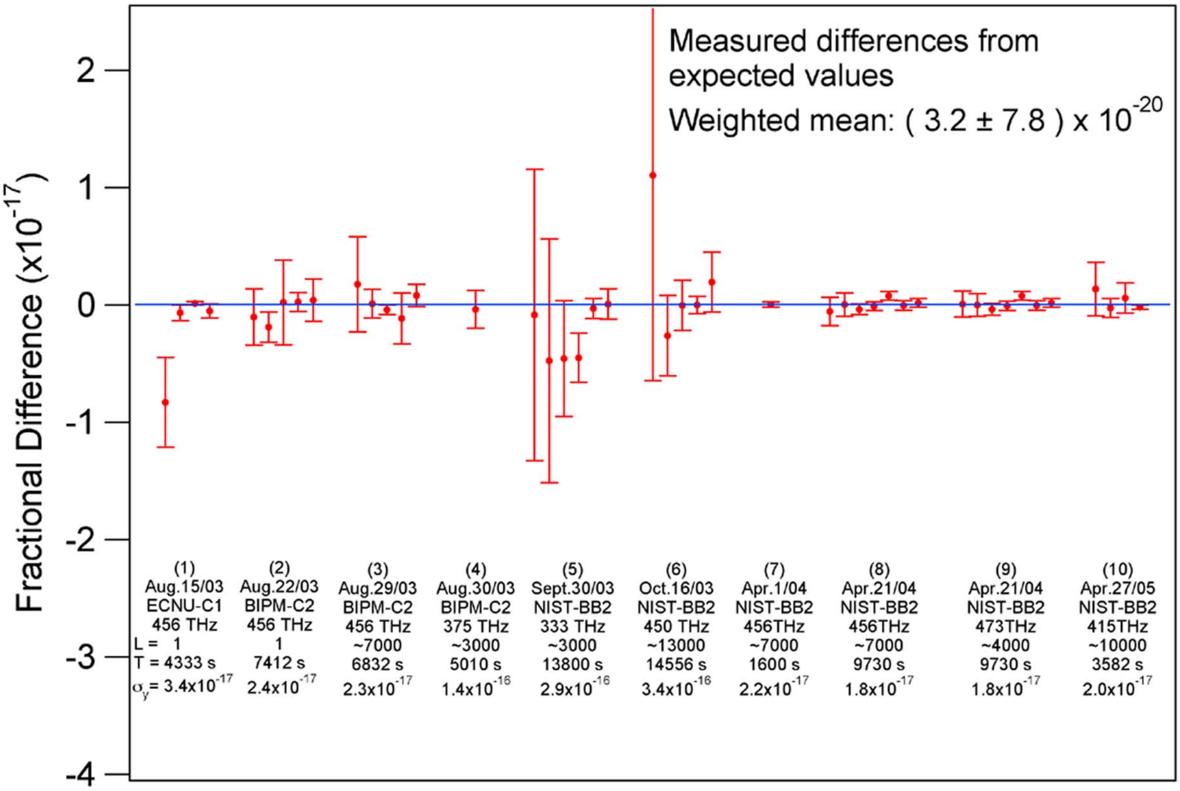

Fig. 5. Summary of optical heterodyne measurements of three FLFCs (NIST-BB2, BIPM-C2, ECNU-C1) compared to NIST-BB1 in 2003, 2004, and 2005. Below each of the ten measurements, we provide the measurement date, the FLFC compared to NIST-BB1, the frequency at which the comparison was made, the number of comb lines compared (L), the total averaging time (T), and the Allan deviation for 1 s averaging time (σ_y). The average for the first two measurements $(L = 1)$ is $(4.5 \pm 26) \times 10^{-20}$, while the average of all other measurements $(L > 3000)$ is $(2.6 \pm 8.8) \times 10^{-20}$.

N being a large number, near 5×10^5, makes the optical heterodyne detection a very efficient way to test how precisely the FLFC can be controlled by the optical phase-lock systems. Therefore, using (1) — (4), $\Delta f_{\rm rep}$ and Δf can be determined by $f_{\rm ceo1}, f_{\rm ceo2}, f_{\rm b1}, f_{\rm b2}, N_L$, and N precisely, and the expected values of $\Delta f_{\rm rep}$ or Δf can be compared to the measured values. This makes possible high-precision tests of the spectral purity and intrinsic noise of the two FLFCs themselves.

In most cases of optical heterodyne comparisons, we required $f_{\rm rep1} = f_{\rm rep2}$. This allows the use of groups of lines from each of the two combs to generate the frequency difference signal. In this case, the expected beat frequency between the two FLFCs can be written as

$$\Delta f = f_{\rm ceo1} - f_{\rm ceo2}. \qquad (5)$$

When the relative phase between the optical pulse trains from the two FLFCs is set to zero (i.e., the pulses from each FLFC reach the detector at the same time), all modes are appropriately synchronized to generate a strong beat signal with signal-to-noise ratio as high as 60 dB within a 300-kHz bandwidth [7].

In a few cases of comparisons, with $f_{\rm rep1} \approx f_{\rm rep2}$ but not necessarily equal, we can compare the frequencies of single lines adjacent to mode N_L from each of the two combs. This method does not require time synchronization between the optical pulse trains from the two FLFCs [23].

Fig. 5 shows the relative frequency difference between the measurements of Δf and the expected value for all the comparisons made by optical heterodyne detection in different optical regions over the period of August 2003-April 2005. Each group of points represents measurements taken over thousands seconds on the designated day at wavelengths ranging from 633 to 900 nm. In Fig. 5, we list the frequency at which the comparison occurred and the approximate number of modes involved, both of which are determined by the center wavelength and bandwidth of the optical bandpass filter shown in Fig. 4. Using standard statistical methods (see for example [32]), we combined the data from 10 measurements to calculate the weighted mean. The result is equal to 3.2×10^{-20}, with an uncertainty of 7.8×10^{-20}, corresponding to a 95% confidence level determined from a χ^2 analysis. Thus, we measure no difference between the comb frequencies with an uncertainty at the level of 10^{-19}. We have also separated the average fractional differences by the type of measurement ($L = 1$ and $L > 3000$), and the results are quoted in the caption of Fig. 5.

B. Comparison of $f_{\rm rep}$ of Optical Pulse Trains by Nonlinear Cross Correlation

Fig 6(b) shows the configuration for the comparison of pulse trains by nonlinear cross correlation [33]. This technique effectively measures relative fluctuations in the arrival times of the two optically referenced pulses trains. Since $f_{\rm rep}$ is phase-coherently linked to the optical frequencies of the FLFC, there is some redundancy between this comparison and that described in Section III-A. However, one could envision a situation where the elements of the comb spectrum are fixed at the frequencies

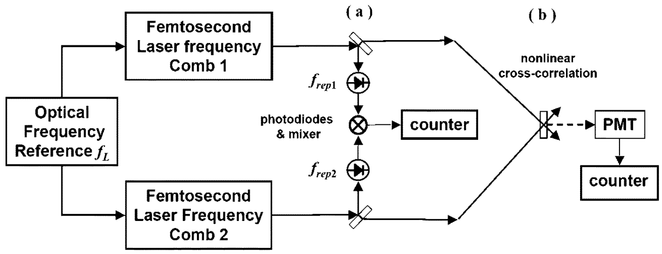

Fig. 6. Experimental setup for the comparison of repetition rates of two FLFCs (a) by photo diode detection and RF mixer (b) by nonlinear cross-correlation of the optical pulse trains.

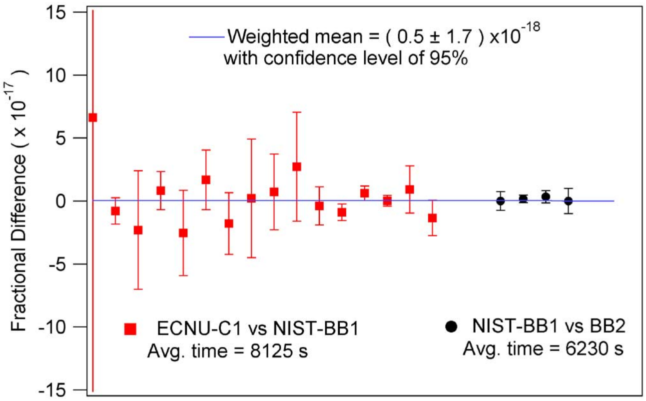

Fig. 7. Results of optical pulse train comparisons that use nonlinear cross-correlation.

given by (2) and (3), but the amplitude of the comb elements fluctuate. Such spectral amplitude fluctuations, coupled with the dispersive elements in each of the FLFC setups, could potentially lead to excess jitter noise and uncertainty in the arrival time of the pulse trains.

Optical pulse trains from two separate FLFCs cross in a beta-barium borate (BBO) nonlinear crystal. When pulses from each laser arrive synchronously at the crystal, the sum-frequency signal is generated. This sum-frequency pulse train bisects the angle between the two crossed pulse trains. The repetition rate of the sum-frequency pulse train can be written as

$$f_{\text{sum}} = |f_{\text{rep1}} - f_{\text{rep2}}| = \Delta f_{\text{rep}}. \tag{6}$$

A photo-multiplier-tube (PMT) with a UV transmitting filter is used to detect f_{sum} which is then measured by a digital counter. The measured data were compared with the expected value given by (1). The FLFCs of NIST-BB2 and ECNU-C1 were compared with NIST-BB1, and Fig. 7 shows the fractional difference between experimental data and the expected value. The calculated weighted mean of 14 355 s of data is equal to 0.5×10^{-18}, with an uncertainty of 1.7×10^{-18}, corresponding to a 95% confidence level.

C. Comparison of Repetition Rate by Photodiode Detection

Many applications of FLFCs, such as the comparison of optical and microwave frequency standards, require the generation of an electronic microwave signal. In such a case, the optically referenced pulse train at f_{rep} is converted to an electronic signal using a high-speed photodiode. The generated photocurrent pulses also consist of a comb of frequencies at harmonics of f_{rep}, extending up to the nominal bandwidth of the photodiode (see Fig. 2). It has previously been shown that the process of photodetection can add excess phase noise via the conversion of amplitude noise to phase noise [34], so it is important to investigate the possible influence of such noise on the uncertainty of f_{rep}.

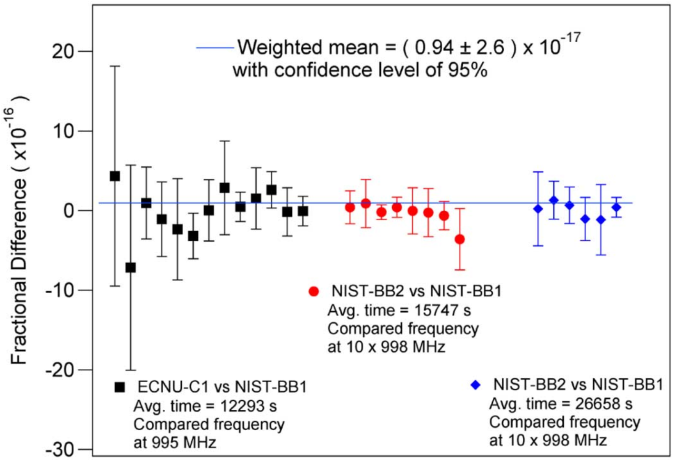

Fig. 8. Results of repetition rate comparisons that use photodiode detection and RF mixing.

The configuration for the comparison of repetition rates by photodiode detection is shown Fig. 6(a). In the comparison between ECNU-C1 and NIST-BB1, the optical pulses were detected on free-space photodiodes. In order to reduce noise associated with beam pointing fluctuations, fiber-coupled photodiodes were used to detect the repetition rate for the comparisons between NIST-BB2 and NIST-BB1 [9], [10]. Two repetition rate signals were filtered and amplified at their 10th harmonics near 10 GHz, which were then mixed to generate the difference of repetition rate (typically 1 kHz or less) between the compared FLFCs. The difference in repetition rates was measured by a digital counter. Fig. 8 shows the fractional difference between the experimental data and the expected values. The calculated weighted mean for the photodetected microwave signals is 0.17×10^{-17}, with an uncertainty of 2.6×10^{-17}, corresponding to a 95% confidence level determined from a χ^2 analysis.

IV. SUMMARY AND DISCUSSION

Table I summarizes the comparative measurements using the three different methods described above. As a general conclusion, the results presented here show that an FLFC based on a mode-locked Ti:sapphire femtosecond laser possesses sufficiently low residual noise to support the best present-day optical and microwave frequency standards. The fractional noise of the FLFC is lowest for measurements performed in the optical domain and increases when one moves to the microwave domain (i.e., f_{rep}), as shown in Fig. 9. The ultimate uncertainty

TABLE I
SUMMARY OF COMPARISONS OF OPTICALLY REFERENCED FEMTOSECOND LASER FREQUENCY COMBS

Compared Systems	Method of Comparison	Compared Frequency	Averaging Time	Allan Deviation (1 s)	Relative Uncertainty
NIST-BB1, NIST-BB2, BIPM-C2, ECNU-C1	Optical Heterodyne	333-473 THz	76 585 s	2×10^{-17}	7.8×10^{-20}
NIST-BB1, NIST-BB2, ECNU-C1	Non-linear Cross-correlation	998 MHz	14 355 s	2×10^{-15}	1.7×10^{-18}
NIST-BB1, NIST-BB2 ECNU-C1	Photo-diode Detection	10×998 MHz & 998 MHz	54 698 s	2×10^{-15}	2.6×10^{-17}

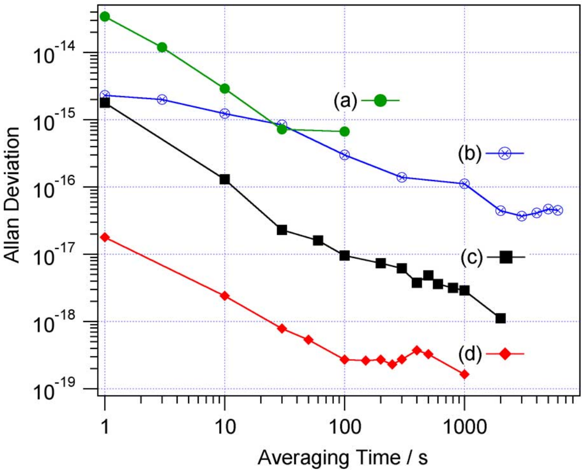

Fig. 9. Relative Allan deviation of the various types of comparisons. (a) Photodiode detection for repetition rate comparison at 995 MHz in the case of ECNU-C1 versus NIST-BB1. (b) Fiber coupled photodiode detection for repetition rate comparison at 10×998 MHz in the case of NIST-BB1 versus NIST-BB2. (c) Comparison of optical pulse train using nonlinear cross-correlation; the data from 1–60 s are the measurements of ECNU-C1 versus NIST-BB1 and the data after 100 s are the measurements of NIST-BB1 versus NIST-BB2 (d) optical heterodyne detection for the comparison of comb line position at 456 and 473 THz.

of an optical or microwave comparison made in a specific averaging time will depend on the short-term instability. The instability (Allan deviation) for the measurements in optical domain can begin near 1×10^{-17} at 1 s and averages down to the 10^{-19} range in a few thousand seconds. For the optical-to-microwave conversion, using nonlinear cross-correlation, the Allan deviation follows the similar behavior, although with instability approximately two orders at magnitude higher. This is due primarily to the increased relative phase noise in the nonlinear cross-correlation when compared to the optical heterodyne method.

Considering the comparison of the frequency position of the optical modes of the FLFC, these results indicate that the FLFC can transfer the properties of an optical frequency standard to other regions of the optical spectra with relative frequency uncertainty below 1×10^{-19}. At this point, we believe the uncertainty of the FLFC in this respect is limited by environmental perturbations. This is supported in part by the data of Fig. 5, which show significantly less scatter for the data of the later experiments (2004 and 2005), when compared to the experiments of 2003 (points 1–6). The main improvements between these two periods result from enclosing the light beam paths and FLFCs in covered boxes, and the arrangement of the optical paths to have better common path rejection and improved immunity to mechanical and thermal fluctuations. The results of these comparisons demonstrate that the FLFC can be reliable tools for the comparison of high performance optical frequency standards.

The comparison of the optical pulse trains shows that the FLFC can transfer an optical frequency to the femtosecond optical pulse train (at frequency f_{rep}) with relative frequency uncertainty below 2×10^{-18}. Since the cross correlation measurement is based on a nonlinear optical process, the intensity of the sum-frequency light, as well as the counting of this sum-frequency signal, is sensitive to the power of the two FLFCs. This provides a means by which amplitude fluctuations can be misinterpreted as phase or timing noise. The present results could be improved with optical power control and lower noise fast photo detection of the sum-frequency pulse train.

The process of converting the pulse train of the optically referenced FLFC to an electronic microwave signal has additional frequency noise associated with it. Nonetheless, our results demonstrate that this can be achieved with relative uncertainty as low as 2.6×10^{-17}, when the microwave signal is generated by the photodetection of the optical pulse train (or its harmonics). At present, we believe this uncertainty is likely limited by amplitude to phase noise conversion in the photodetection process [34]. Not only does photodetection add noise on short (i.e., 1 s) time scales, but it is evident from Fig. 9(b) that the instability of the photodetected signal deviates from the expected $1/\tau$ averaging obtained up to 100 s in the cross-correlation [Fig. 9(c)] and optical heterodyne [Fig. 9(d)] experiments. We can speculate that this is the result of slower power-, temperature-, or other and environment-driven fluctuations in the photodiodes and electronics. In any case these results show that self-referenced FLFCs can serve as frequency synthesizers from optical to microwave frequencies with unprecedented reproducibility and residual frequency noise.

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Long-Sheng Ma was born in 1941 in Shanghai, China. He graduated from East China Normal University in electronics and physics in 1963.

He studied laser propagation in atmosphere and monitoring of air pollution until 1980. Since 1981, his research has been in the field of ultrasensitive and high resolution laser spectroscopy, laser frequency stabilization, and optical frequency measurements. He was a Visiting Professor at the Joint Institute for Laboratory Astrophysics (JILA), University of Colorado, Boulder, and a Guest Scientist at the Institute for Laser-Physics, Hamburg University, Hamburg, Germany, and National Institute of Standards and Technology (NIST), Boulder, CO. He was a Visiting Scientist as a Senior Research Fellow at the Bureau International des Poids et Mesures (BIPM), Sevres, France. His permanent position is as Professor of Physics Department, East China Normal University, Shanghai, China.

Zhiyi Bi was born in Shanghai, China, in 1956. He received the B.S. degree in physics from Shanghai University of Technology, Shanghai, China, in 1982, and the M.S. degree in physics from East China Normal University, Shanghai, China, in 1986.

He is currently a Professor in the Department of Physics at East China Normal University. His research interests include precision control of optical fields and ultrasensitive laser spectroscopy.

Albrecht Bartels was born in Orsoy, Germany, in 1972. He received the diploma and the Ph.D. in physics in 1997 and 2000 from RWTH Aachen University, Germany. In his Ph.D. work, he built the first Ti: sapphire femtosecond laser with repetition rates above 1 GHz.

During postdoctoral positions at RWTH Aachen University, Germany (2000–2001) and the Time and Frequency Division of the National Institute of Standards and Technology at Boulder, CO (2001–2004), he explored possibilities to use gigahertz repetition rate femtosecond lasers for the generation of ultrastable optical frequency combs. He is currently CEO of Gigaoptics GmbH, Konstanz, Germany, and has a teaching position at University of Konstanz. His current research interest are ultrafast lasers, optical frequency combs, and novel methods for time-domain terahertz and pump-probe spectroscopy.

Kyoungsik Kim received the B.S. and M.S. degrees in physics from Seoul National University, Seoul, Korea, in 1992 and 1994, respectively. He received the Ph.D. degree in applied physics from the University of Michigan, Ann Arbor, in 2004, where he worked in the area of femtosecond laser spectroscopy of semiconductor quantum dots.

He was a Full-Time Lecturer of physics at the Korean Air Force Academy, Cheongwon, Korea, for military service from 1994 to 1997. His postdoctoral research was performed at National Institute of Standards and Technology, Boulder, CO, during 2004–2005. He is currently an Assistant Professor in the School of Mechanical Engineering, Yonsei University, Seoul, Korea.

Lennart Robertsson was born in Vänersborg, Sweden, in 1953. He received the B.Sc. and Ph.D. degrees from the University of Gothenburg, Gothenburg, Sweden, in 1978 and 1984, respectively.

In 1984, he joined the Institute for Optical Research, Stockholm, Sweden, to work with interferometric methods for optical testing and precision machining techniques for the production of optical surfaces. Since 1988, he has been with the Bureau International des Poids et Mesures (BIPM), Sevres, France, where he is involved in optical frequency measurements using femtosecond comb techniques and stabilised lasers.

Massimo Zucco was born in Turin, Italy, in 1964. He received a degree in physics from the University of Turin, Turin, Italy, in 1990 and the Ph.D. degree in metrology from the Polytechnic of Turin, Turin, Italy, in 1995, respectively.

He worked for two years at the National Physics Laboratory (NPL) as a Research Fellow on the realization of the caesium fountain, then for four years at the Bureau International des Poids et Mesures (BIPM), Sevres, France, on absolute optical frequency measurements using femtosecond comb techniques and on laser metrology. He now has a permanent position at the National Institute of Metrological Research (INRIM), Turin, Italy.

Robert S. Windeler received the B.S. degree in chemical engineering from the University of Delaware, Newark, in 1990, and the M.S. and Ph.D. degrees in chemical engineering from the University of California, Los Angeles, in 1992 and 1995, respectively.

He became a Member of Technical Staff at Bell Laboratories, Lucent Technologies (now OFS Laboratories), Murray Hill, NJ, in 1995 and is a member of the Optical Fiber Research Department where he has extensive experience in fiber fabrication and design. His areas of expertise include MCVD, modified MCVD, and solution doping which he has used to produce world class specialty fibers. His current research efforts involve fabrication and design of Er–Yb double-clad fiber for high-power amplifiers and microstructure air-silica fibers. His microstructure fibers were used to demonstrate for the first time anomalous dispersion below 1290 nm, zero group-velocity dispersion as low as 765 nm, and many nonlinear interactions in the near visible region including soliton propagation, self-phase modulation, pulse compression, and ultrabroad-band continuum generation. He holds six patents and has over ten pending patents on production and use of optical fiber.

Guido Wilpers received thee German Physik-Diplom from the Gerhard-Mercator-Universitaet Gesamthochschule Duisburg, after carrying out his Diplomarbeit on a diagnostic for analysing neutral exhaust gases from the divertor of the JET experimental fusion reactor in Culham, U.K. He received the Ph.D. degree (Dr. rer. nat.) from the University of Hannover, Hannover, Germany, for his work on the setup, characterization, and frequency measurement of an optical frequency standard with millikelvin neutral calcium atoms at the PTB in Braunschweig, Germany.

He then moved on to work as a postdoc at National Institute of Standards and Technology (NIST), Boulder, CO, on a similar frequency standard on the implementation of microkelvin calcium atoms and frequency measurements of the optical clock transition. He is presently participating as a Strategic Research Fellow at the National Physical Laboratory (NPL), Teddington, U.K., in establishing quantum information processing capability based on strontium ions in segmented linear RF-Paul traps and the development of entanglement based measurements for ion optical frequency standards.

Chris Oates was born in Glendale, CA, in 1962. He received the B.S. degree in physics from Stanford University, Stanford, CA, in 1984, and the Ph.D. degree from the University of Colorado, Boulder, in 1995.

After spending two years teaching with Peace Corps, he attended the University of Colorado, where his research focused on precision spectroscopy of laser-cooled atoms. In 1995, he joined the National Institute of Standards and Technology, Boulder, CO, where he works on optical frequency standards based on neutral atoms.

Leo Hollberg received the B.S. degree in physics from Stanford University, Stanford, CA, in 1976, and the Ph.D. degree in physics at the University of Colorado, Boulder, for research in high-resolution laser spectroscopy done under the supervision of J. Hall at the Joint Institute for Laboratory Astrophysics (JILA), Boulder, CO.

Most of 1984 and 1985 were spent at AT&T Bell Laboratories as a Postdoctoral Researcher working with S. Chu on laser cooling and trapping of atoms, and with R. Slusher on squeezed states of light. Since then, he has been at the National Institute of Standards and Technology (NIST), Boulder, CO, doing research on high-resolution spectroscopy of laser-cooled and -trapped atoms, the development of semiconductor lasers for scientific and technical applications, optical coherence effects of driven multilevel atoms, chip-scale-atomic-clocks, optical frequency standards, optical frequency combs and optical atomic clocks. His areas of expertise include frequency stabilized lasers with ultranarrow linewidths and high resolution optical spectroscopy and optical frequency standards. Much of this research is done in collaboration with his NIST colleagues and with scientists from around the world. Leo is currently the group leader of the Optical Frequency Measurements group in the Time and Frequency Division, NIST, Boulder.

Scott A. Diddams (M'99) received the B.A. degree in physics from Bethel College, St. Paul, MN, in 1989 and the Ph.D. degree in optical science from the University of New Mexico, Albuquerque, in 1996.

Between 1996 and 2000, he did postdoctoral work at the Joint Institute for Laboratory Astrophysics (JILA), Boulder, CO, where he was supported in part by a National Research Council fellowship. While working in the laboratpry of John Hall, he carried out experiments resulting in the first carrier-envelope stabilization of femtosecond laser pulses and direct counting of optical frequencies with octave-spanning femtosecond laser frequency combs. Currently, he works as a physicist in the National Institute of Standards and Technology (NIST), Boulder, CO, where he pursues research within the fields of nonlinear optics, ultrafast lasers and phenomena, and precision spectroscopy and metrology.