

PHYSICS

Optical frequency divider with division uncertainty at the 10^{-21} level

Yuan Yao¹, Yanyi Jiang^{1,2,*}, Hongfu Yu¹, Zhiyi Bi^{1,2} and Longsheng Ma^{1,2,*}**ABSTRACT**

Optical clocks with unprecedented accuracy of 10^{-18} promise innovations in many research areas. Their applications rely to a large extent on the ability of precisely converting the frequency from one optical clock to another, or particularly to the frequencies in the fiber telecom band for long-distance transmission. This report demonstrates a low-noise, high-precision optical frequency divider, which realizes accurate optical frequency conversion and enables precise measurement of optical frequency ratios. By measuring against the frequency ratio between the fundamental and the second harmonic of a 1064-nm laser instead of a second copy of the same system, we demonstrate that the optical frequency divider has a fractional frequency division instability of 6×10^{-19} at 1 s and a fractional frequency division uncertainty of 1.4×10^{-21} . The remarkable numbers can support frequency division of the best optical clocks in the world without frequency-conversion-caused degradation of their performance.

Keywords: optical frequency comb, optical atomic clock, precision measurement, frequency metrology

INTRODUCTION

Recent progress in optical atomic clocks demonstrates record fractional frequency instability and uncertainty at the 10^{-18} level [1–4]. The unprecedented accuracy is fostering a revolution in science and technology [5]. Using optical clocks, searches for possible variations of fundamental constants are carried out in laboratories by precisely measuring the frequency ratios of different atomic transitions of optical clocks over time [6–8]. In relativistic geodesy, long-distance geopotential difference will be accurately measured by comparing the frequencies of remotely located optical clocks linked with optical fibers [5,9–11], where the frequencies of optical clocks have to be accurately converted to the fiber telecom band for long-distance transmission. In metrology, the fundamental unit for time, the *second*, in the International System of Units (SI) will be redefined based on optical atomic clocks. Frequency comparisons between optical clocks based on different atom species have to be performed in order to affirm the agreement between optical clocks with uncertainty beyond the current SI second, as well as to demonstrate the frequency reproducibility of optical clocks [12]. Moreover, in atomic and

molecular precision spectroscopy, hopes are high that accurate and stable clock light can be transferred to wider spectral range [13,14].

All those applications rely on accurate frequency ratio measurement between spectrally separated optical clocks or frequency conversion of optical clocks. Optical frequency combs [15,16] are usually employed to link the frequencies of optical clocks. By synchronously counting the frequency of the repetition rate (f_r) and the carrier-envelope offset frequency (f_0) of a comb, as well as the beating frequencies between optical clocks and their nearby comb teeth relative to a hydrogen maser, frequency ratios between optical clocks were measured [6,11,17,18]. To investigate the limit of optical frequency ratio measurement determined by the measuring setups instead of optical clocks, Johnson *et al.* measured a same optical frequency ratio with two setups based on different types of optical frequency combs. They use synchronous counting referenced to a common hydrogen maser and the transfer oscillator scheme [19,20] to remove the comb frequency noise, and demonstrate the systematic instability and agreement of two setups to be 2.8×10^{-16} at 1-s averaging time and 3×10^{-21} , accordingly [21]. Supported by

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the development of the state-of-the-art ultrastable lasers with a frequency instability of 10^{-17} and better in the near future [22–25] and the efforts on using correlated atomic samples to overcome the standard quantum limit [26–28], the frequency stability of optical atomic clocks is bound to set a new record, pushing for an even more precise way to quickly determine the frequency ratios.

Here we report a low-noise, high-precision optical frequency divider (OFD) in the visible and near infrared regions, capable of linking all the present optical clocks aided by the nonlinear optical second-harmonic generation. Compared with the previous optical frequency dividers [29–31], the OFD described here can accurately divide the frequency of a laser with a preset arbitrary division ratio to another wavelength or to frequencies in the fiber telecom band [32] or in the microwave region [29,30] for a wide range of applications. It is convenient to use providing arbitrary-wavelength ultrastable laser with high precision. By using our OFD, the frequency ratios between optical clocks can be precisely determined directly from the divisor of the OFD. In particular, using an OFD like ours, the frequency of a high-performance clock laser can be converted to another at a different wavelength, which improves the frequency ratio measurement by partially cancelling out the laser frequency noise in synchroniza-

tion operation of optical clocks [18]. By comparing against the frequency ratio between the fundamental and the second harmonic of a 1064-nm laser, the fractional frequency instability of the OFD output light relative to the input light is demonstrated to be 6×10^{-19} at 1 s and 2×10^{-20} at 1000 s. The fractional uncertainty of the OFD in optical frequency division is characterized to be 1.4×10^{-21} . The OFD thus can support frequency division of the most stable lasers [22,33–36] and the most accurate optical clocks in the world without degrading the performance, and enables precision measurements at the 10^{-21} level.

RESULTS

Experimental realization

The output light frequency of our OFD is directly related to the input light frequency with a precise divisor R , $f_{\text{out}} = f_{\text{in}}/R$. The experimental schematic for realizing the OFD is shown in Fig. 1. An output laser (f_{out}) is phase-locked to the input light (f_{in}) via an optical frequency comb (see Methods). The comb is based on a Ti:sapphire mode-locked femtosecond laser with a repetition rate of 800 MHz. The frequency of the N th comb tooth is $f_N = Nf_r + f_0$. To reduce the comb frequency noise, the comb is optically referenced to f_{in} by phase-locking the N_1 th comb tooth to f_{in} and by stabilizing f_0 to a stable RF [37]. The beat signal of f_{b1} (f_{b2}) between f_{in} (f_{out}) and its nearby N_1 th (N_2 th) comb tooth is generated on a photo detector. A tunable radio frequency (RF) signal f_{tune} is mixed with f_{b2} on a double balanced mixer (DBM) for precise tuning of f_{out} as well as R of the OFD conveniently. To remove the residual frequency noise of the comb, f_0 and f_r are subtracted based on the transfer oscillator scheme [19,20] to make the error signal Δ for phase-locking f_{out} to f_{in} independent of f_0 and f_r . Consequently, the resulting error signal is $\Delta = f_{\text{in}}/M_1 - (f_{\text{out}} - f_{\text{tune}})/M_2$, where M_1 and M_2 are the divisors of two direct digital synthesizer (DDS), which are chosen to meet $M_1/M_2 = N_1/N_2$ in order to remove f_r from Δ . Then, the signal Δ is sent to a servo to lock the frequency of the output laser f_{out} to f_{in} as $f_{\text{out}} = (M_2/M_1) \times f_{\text{in}} + f_{\text{tune}}$.

In order to realize a real optical frequency division with a precise division ratio, here, f_{tune} has to be related to f_{in} only. To achieve this goal, the beat notes of f_{b1} and f_{b1}^* between f_{in} and its two neighboring comb teeth of the same comb are detected on a photo detector. After removing f_0 and f_r using DBMs and DDSs in a similar manner as described above, the resulting beat signal is directly derived from f_{in} as $f_{\text{tune}} = f_{\text{in}}/K$, where K is a number depending on the settings of the DDSs and an RF synthesizer (see

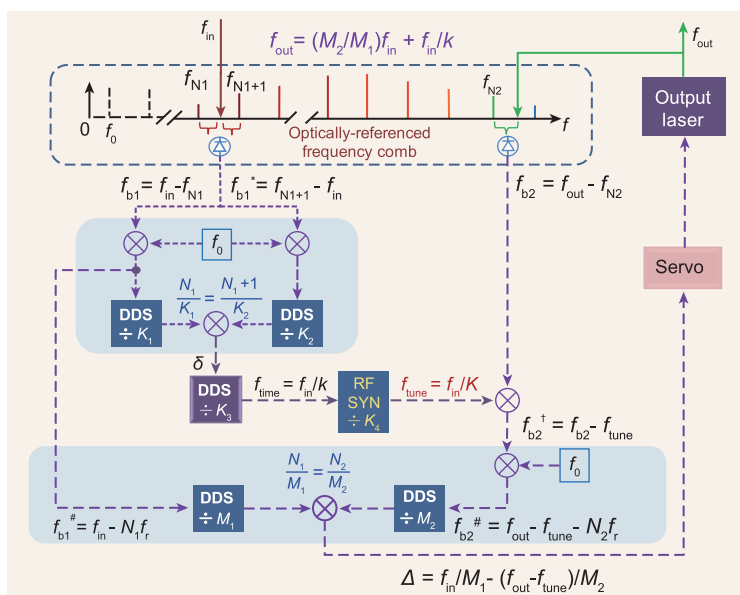


Figure 1. Experimental realization. f_{out} is related to f_{in} as $f_{\text{out}} = f_{\text{in}}/R$ with an optically referenced frequency comb. The beating signals of f_{b1} and f_{b2} between f_{in} , f_{out} and their nearby comb teeth are detected to generate an error signal Δ for phase-locking. The residual frequency noise of f_0 is subtracted from the beating signals on double balance mixers (DBM), and the residual frequency noise of f_r is removed with direct digital synthesizers (DDS) and a DBM based on the transfer oscillator scheme. A tunable RF signal f_{tune} synthesized from f_{in} is used for precise adjustment of f_{out} and R . RF SYN denotes RF synthesizer.

Methods). Benefiting from this self-referenced RF signal, f_{out} is directly divided from f_{in} with a ratio $R = 1/(M_2/M_1 + 1/K)$. If f_{tune} is set with a resolution of $1 \mu\text{Hz}$, the ratio R can be set at the 21th decimal place to an arbitrary pre-determined value when both f_{in} and f_{out} are within the spectrum of the comb.

Optical frequency division noise and uncertainty

To characterize the performance of the OFD, we measured the divisor R of the OFD against the frequency ratio between the fundamental and the second harmonic of a 1064-nm laser (see Methods) instead of comparing against a second copy of the identical OFD. The second-harmonic generation re-

alizes exactly an OFD with divisor at 0.5 to the precision we demonstrate.

The schematic diagram of the measurement is shown in Fig. 2a. We used the OFD to realize optical frequency division as $f_{532} = f_{1064-1}/R_x$, where f_{1064-1} is the frequency of a cavity-stabilized laser at 1064 nm [38] and f_{532} is the frequency of the light second-harmonic generated from an independent 1064-nm laser (f_{1064-2}) in a nonlinear crystal. The laser frequency f_{532} is tuned by adjusting the voltage of a piezo transducer (PZT) inside the laser cavity of f_{1064-2} . The divisor R_x is set arbitrarily as 0.500 000 053 261 644 522 938. Both the laser light of f_{532} and f_{1064-2} propagate collinearly and combine with the optically referenced comb light on a beam splitter (BS) to reduce the effect of light path fluctuation, as shown in Fig. 2b. To further eliminate the light-path-fluctuation-induced frequency noise, the light from f_{1064-1} co-propagates with the comb light through a piece of photonic crystal fiber (PCF) to a photo detector (PD₁). Besides, all the optics are well sealed in a box to reduce the fluctuation of the light path due to airflow turbulence.

Then, the beating frequency f_b between f_{1064-1} and f_{1064-2} was measured on a frequency counter. Profiting from the self-referenced time base at nearly 10 MHz for the counter which is down-converted from f_{1064-1} as $f_{\text{time}} = f_{1064-1}/k$ with a preset ratio $k = K_3/(1/K_1 - 1/K_2)$, here K_1 , K_2 and K_3 are the divisors of the DDSs (see Fig. 1 and Methods). The value of R_x can be determined from the equation

$$f_b = f_{1064-1} - f_{1064-2} = f_{1064-1} - \frac{f_{1064-1}}{2R_x} = \frac{30.1 \times 10^6 - A}{10^7} \times \frac{f_{1064-1}}{k} = f_{1064-1} - \frac{f_{1064-1}}{2R_x} \quad (1)$$

where A is the reading number in hertz on the counter.

We measured the value of R_x on nine different days over 15 d (a total measurement time of 105 000 s). The fractional instability of the measured R_x is shown in Fig. 3a with red dots. The low instability of 6×10^{-19} at 1-s averaging time benefits from both the optically referenced frequency comb and the elimination of light path fluctuation. The measured noise of the OFD is from servo systems, all the DDSs and residual light path fluctuation. The measured instability is close to $4 \times 10^{-19}/\sqrt{\tau}$ (τ is the averaging time) when f_{1064-1} is directly phase-locked to f_{1064-2} without using an optical frequency comb (blue squares), where the noise comes from a servo system and light path fluctuation. It implies that, during optical frequency division, the relative frequency instability between the output and input of the OFD is more than two orders of magnitude better than

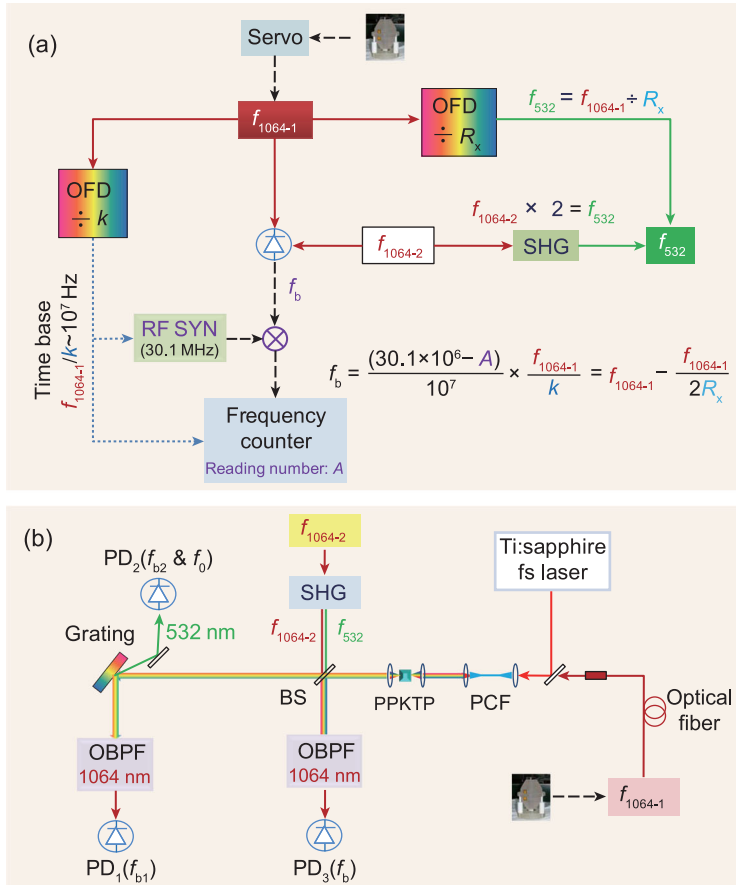


Figure 2. Diagram of performance measurement. (a) Schematic diagram. (b) Experimental diagram. We used the OFD to connect the optical frequencies of a cavity-stabilized laser at 1064 nm (f_{1064-1}) and the light with frequency f_{532} which is second-harmonic generated from an independent 1064-nm laser (f_{1064-2}). As a result, $f_{532} = f_{1064-1}/R_x$. The uncertainty of R_x is obtained by measuring the beating frequency (f_b) between f_{1064-1} and f_{1064-2} on an RF frequency counter with a self-referenced time base of f_{1064-1}/k . fs laser, femtosecond laser; PCF, photonic crystal fiber; PPKTP, periodically poled KTP; SHG, second-harmonic generation; BS, beam splitter; PD, photo detector; OBPF, optical band pass filter; f_{b1} , the beat note between f_{1064-1} and the comb; f_{b2} , the beat note between f_{532} and the comb; f_0 , the carrier-envelope offset frequency of the comb.

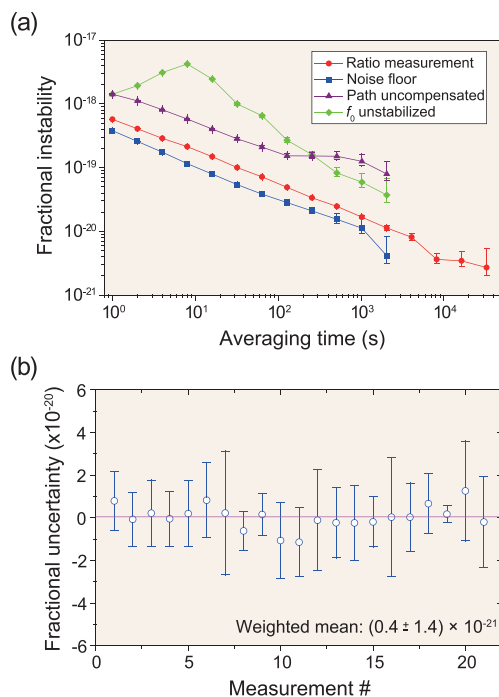


Figure 3. Division noise and uncertainty measurement. (a) Fractional Allan deviation of the frequency ratio between f_{1064-1} and f_{1064-2} when f_{1064-2} is phase-locked to f_{1064-1} without the OFD (noise floor, blue squares) and when f_{1064-2} is frequency divided from f_{1064-1} by the second-harmonic generation (SHG) and an OFD (red dots). The purple triangles and green diamonds are the instability of the frequency ratio between two 1064-nm lasers connected with SHG and OFD when light path is not optimized and f_0 is free running accordingly. (b) Measured R_x deviated from the setting value on nine different days over 15 d. Each data point results from averaging over a roughly 5000-s measurement time. The error bar for each data point is the standard deviation of five mean values. Each mean value is averaged over 1000-s measurement time. The solid pink line denotes the mean value of the total 21 sets of data.

that of the most stable lasers [22,33–36], adding a negligible noise onto the output of the OFD. When the light path in this measurement is not optimized, for instance, the light beams of f_{532} and f_{1064-2} propagate separately before beating against the comb light, it becomes extremely challenging for the instability to average down to below 10^{-19} mainly due to the effect of vibration and temperature fluctuation on tens of centimeters of free-space propagation, as shown with purple triangles in Fig. 3a.

Figure 3b shows the fractional uncertainty of R_x deviated from the OFD setting value. Each piece of data is averaged over a 5000-s measurement time. Using standard statistical methods, we combine 21 sets of data to calculate the weighted mean of 0.4×10^{-21} and a weighted fractional uncertainty to be 1.4×10^{-21} , corresponding to a 99% confidence level determined from a χ^2 analysis [39]. The divi-

sion uncertainty induced by the OFD we demonstrate is therefore three orders of magnitude better than the most accurate optical clocks [3,4].

Transfer oscillator scheme with optically referenced frequency comb

One of the highly acclaimed merits for the transfer oscillator scheme is its immunity to comb frequency noise. When suitably adopted, it is sometimes considered unnecessary to phase-lock the comb to f_{1064-1} . However, due to the limited response bandwidth of the DDSs, the rapid temporally varying signals sent to the DDSs affect the performance of the system. To be sure of the relevant limit, we measured the frequency instability of the beat signal f_b between two 1064-nm lasers (f_{1064-1} and f_{1064-2}), whose frequencies are linked by the OFD as $f_{1064-2} = f_{1064-1}/R$, when a comb tooth is phase-locked to f_{1064-1} but with f_0 free running. The drift rate of f_0 is within 20 kHz/s. The results are shown in Fig. 2b with green diamonds. The fractional instability of R deteriorated to 1×10^{-18} at 1 s and 4×10^{-18} at 10 s. This is way short of the performance levels we report for our OFD. Therefore, in our experimental setup, we used an optically referenced frequency comb, in which f_0 and f_r were stabilized to fluctuate within ± 1 mHz measured with a gate time of 1 s. An additional benefit for an optically referenced frequency comb is that it ensures that all the signals stay within the bandwidth of RF filters.

We further checked that, if one of the divisors of the DDSs (K_1, K_2, M_1, M_2) was set incorrectly, we could easily observe the error by counting the beating frequency f_b between two lasers linked with our OFD when either f_0 or f_r was tuned. Figure 4 shows that f_b fluctuates over time when the comb offset frequency f_0 is swept with an amplitude of 3 kHz and a period of 10 s on purpose. If a divisor of the DDSs for subtraction f_r is set incorrectly, for example, $M_1/M_2 = (N_1 + 1)/N_2$, the beating frequency f is found to fluctuate with a period of 10 s as well, as shown by the blue line in Fig. 4b. Since one tooth of the optically referenced frequency comb is frequency-stabilized to f_{1064-1} , f_r changes when sweeping f_0 . The imperfect subtraction of f_r subsequently leads to the frequency fluctuation of f_{1064-2} . Moreover, it causes the laser to lose phase lock frequently. When the divisors of the DDSs are set correctly, on the other hand, as expected, the beating frequency between two lasers are more stable, as shown in Fig. 4a.

CONCLUSION

For the current experimental setup, the divisor range of our OFD in the optical domain was 0.5–1 if the

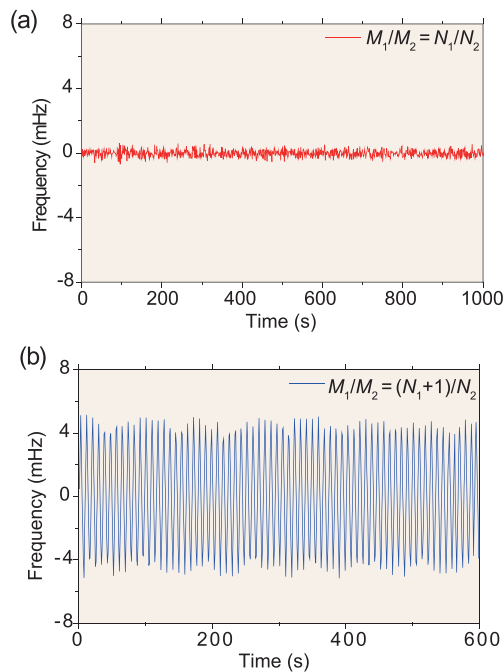


Figure 4. Testing the immunity of comb frequency noise. f_{1064-1} is firstly divided by R_1 to the frequency of a Ti:sapphire continuous wave laser with an OFD, and then the frequency of the Ti:sapphire continuous wave laser is further divided by R_2 to f_{1064-2} with another OFD based on the same comb. The figure shows the frequency fluctuation of the beat signal between f_{1064-1} and f_{1064-2} when the comb offset frequency (f_0) fluctuates with an amplitude of 3 kHz and a period of 10 s on purpose and the divisors of the DDSs for subtraction f_r in one of division steps are set (a) correctly as $M_1/M_2 = N_1/N_2$ and (b) incorrectly as $M_1/M_2 = (N_1+1)/N_2$.

input laser was at $1 \mu\text{m}$, determined by the spectrum of the optical frequency comb, which was in the range of $0.5\text{--}1 \mu\text{m}$. With nonlinear techniques such as second-harmonic generation or optical frequency summing, the divisor of the OFD we demonstrated could be further extended to $0.5\text{--}10$ if the input laser is at $1 \mu\text{m}$, covering the optical wavelength from 0.5 to $10 \mu\text{m}$.

Benefiting from the combination of the optically referenced frequency comb, the self-referenced time base for frequency tuning and counting, the transfer oscillator scheme and careful elimination of the light path fluctuation, an OFD with uncertainty at the 10^{-21} level has been demonstrated. Such a record precision meets many applications of the state-of-the-art optical clocks as well as the next generation of optical clocks [27,40]. With this OFD, we can accurately measure the frequency ratios between optical signals. Meanwhile, it can accurately convert one laser frequency to other laser frequencies, paving the way for optical frequency synthesizers [13,14]. We expect that this type of OFD will be instrumental in precision measurements.

METHODS

Basic operation principle

As shown in Fig. 1, a beat signal between the input laser light (f_{in}) and its nearby comb tooth (f_{N1}) can be written as

$$f_{b1} = f_{\text{in}} - f_{N1} = f_{\text{in}} - f_0 - N_1 f_r. \quad (2)$$

Meanwhile, another beat signal between the output laser (f_{out}) and its nearby comb tooth (f_{N2}) can be written as

$$f_{b2} = f_{\text{out}} - f_{N2} = f_{\text{out}} - f_0 - N_2 f_r, \quad (3)$$

where N_1 and N_2 are integers associated with the particular comb teeth. By measuring f_{in} and f_{out} on a wave meter with an uncertainty of ± 30 MHz, together with the values of the repetition rate f_r and f_0 of the comb measured on frequency counters, N_1 and N_2 can be accurately determined in advance. For the convenience of precise tuning of the output laser frequency f_{out} as well as R of the OFD, f_{b2} is mixed with a tunable RF signal f_{tune} on a DBM. The resulting signal is

$$f_{b2}^\dagger = f_{\text{out}} - f_0 - N_2 f_r - f_{\text{tune}}. \quad (4)$$

The residual frequency noise of f_0 is removed by mixing f_{b1} and f_{b2}^\dagger with f_0 in DBMs. The signal of f_0 is detected by using a collinear self-referencing $1f\text{-}2f$ setup [41], in which the light beams on the red and blue side of the comb spectrum for generating the signal f_0 propagate in a common path to the detector. Therefore, the detected extra frequency noise of f_0 due to the light path fluctuation is negligible. The outputs of the DBMs are sent to two DDSs with divisors of M_1 and M_2 , respectively. Usually, the signal-to-noise ratio (SNR) of a signal input to a DDS is more than 30 dB in a resolution bandwidth (RBW) of 300 kHz. The divisors of the DDSs, M_1 and M_2 , are chosen to satisfy $M_1/M_2 = N_1/N_2$ in order to make the error signal Δ free from f_r . The outputs of the DDSs are compared on a DBM to generate an error signal as

$$\begin{aligned} \Delta &= \frac{f_{b1} + f_0}{M_1} - \frac{f_{b2} + f_0}{M_2} \\ &= \frac{f_{\text{in}} - N_1 f_r}{M_1} - \frac{f_{\text{out}} - N_2 f_r - f_{\text{tune}}}{M_2} \\ &= \frac{f_{\text{in}}}{M_1} - \frac{f_{\text{out}} - f_{\text{tune}}}{M_2}. \end{aligned} \quad (5)$$

Then, the error signal is sent to a servo to adjust the frequency of the output laser to make $\Delta = 0$. Therefore, $f_{\text{out}} = (M_2/M_1)f_{\text{in}} + f_{\text{tune}}$.

In order to set the division ratio precisely, here f_{tune} has to be related to f_{in} only. The beat signals of f_{b1} and f_{b1}^* between f_{in} and the two nearest teeth of the same comb are detected on a photo detector: $f_{b1}^* = f_{N_1+1} - f_{\text{in}} = f_0 + (N_1 + 1)f_r - f_{\text{in}}$. After removing f_0 and f_r using DBMs and DDSs, the resulting signal is directly derived from f_{in} as

$$\begin{aligned} \delta &= \frac{f_{b1} + f_0}{K_1} + \frac{f_{b1}^* - f_0}{K_2} \\ &= \frac{f_{\text{in}} - N_1 f_r}{K_1} + \frac{(N_1 + 1)f_r - f_{\text{in}}}{K_2} \\ &= \frac{f_{\text{in}}}{K_1} - \frac{f_{\text{in}}}{K_2}, \end{aligned} \quad (6)$$

where K_1 and K_2 are the divisors of the DDSs, which are chosen to satisfy $K_1/K_2 = N_1/(N_1 + 1)$. In addition, a DDS with the divisor of K_3 is used to synthesize a self-referenced time base f_{time} at about 10 MHz from δ . Therefore, $f_{\text{time}} = f_{\text{in}}/k$. Here, $k = K_3/(1/K_1 - 1/K_2)$. Using this time base, an RF tuning signal f_{tune} is generated from an RF synthesizer (RF SYN) as $f_{\text{tune}} = f_{\text{in}}/K$, where $K = k \times K_4$ and K_4 is the frequency setting value of RF SYN. Therefore, the frequency of the OFD output light is directly divided from f_{in} with a ratio of $R = 1/(M_2/M_1 + 1/K)$.

Division uncertainty measurement

Laser light at 532 nm (f_{532}) is second-harmonic generated from an independent Nd:YAG laser at 1064 nm (f_{1064-2}) by a nonlinear crystal. Both the 532 nm laser light and 1064 nm laser light propagate collinearly and combine with the optically referenced frequency comb light on BS, as shown in Fig. 2b. The beat note (f_{b2}) between f_{532} and the comb is detected on a photo detector (PD₂), while the beat note (f_{b1}) between f_{1064-1} and the comb is detected on PD₁ with a SNR of 50 dB (RBW = 300 kHz). With the signals of f_{b1} , f_{b2} and f_0 , we use the OFD to divide f_{1064-1} to f_{532} with a setting ratio R_x . In our measurement, R_x is set arbitrarily as 0.500 000 053 261 644 522 938.

The beat note (f_b) between f_{1064-1} and f_{1064-2} is detected on PD₃ with a SNR of 40 dB (RBW = 300 kHz). The frequency of f_b is nearly 30 MHz determined by R_x . Then, f_b is mixed down with an RF signal of 30.1 MHz from an RF synthesizer to nearly 100 kHz, which is filtered in a low pass filter and is counted on a frequency counter (Λ -type, gate time = 1 s). The time base at near 10 MHz (10^7 Hz) for the RF frequency counter and the RF synthesizers is the self-referenced time base of $f_{\text{time}} = f_{1064-1}/k$. Therefore, f_b is related

to f_{1064-1} as

$$f_b = \frac{30.1 \times 10^6 - A}{10^7} \times \frac{f_{1064-1}}{k}, \quad (7)$$

where A is the reading number on the counter. With

$$\begin{aligned} f_b &= f_{1064-1} - f_{1064-2} = f_{1064-1} \\ &\quad - \frac{f_{532}}{2} = f_{1064-1} - \frac{f_{1064-1}}{2R_x}, \end{aligned} \quad (8)$$

the frequency ratio R_x can be obtained as

$$R_x = \frac{1}{2 \left(1 - \frac{30.1 \times 10^6 - A}{k \times 10^7} \right)}. \quad (9)$$

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