

Comb-Resolved Dual-Comb Spectroscopy Stabilized by Free-Running Continuous-Wave Lasers

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We demonstrate dual-comb spectroscopy with relatively phase-locked two frequency combs, instead of frequency combs firmly fixed to the absolute frequency references. By stabilizing two beat frequencies between two mode-locked lasers at different wavelengths observed via free-running continuous-wave (CW) lasers, two combs are tightly phase locked to each other. The frequency noise of the CW lasers barely affects the performance of dual-comb spectroscopy because of the extremely fast common-mode noise rejection. Transform-limited comb-resolved dual-comb spectroscopy with a 6 Hz radio frequency linewidth is demonstrated by the use of Yb-fiber oscillators.

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The output of a mode-locked laser forms hundred-thousand of longitudinal modes with a spacing set by the laser repetition frequency.¹⁾ Various metrological applications such as optical clocks, precision spectroscopies, and stable microwave synthesis have progressed significantly from advances in frequency comb technology.²⁾ Recently, dual-comb spectroscopy³⁾ using two mode-locked lasers with slightly different repetition frequencies has attracted attention as a new spectroscopic tool, and has already been applied to molecular absorption spectroscopy,⁴⁻⁷⁾ distance measurement,⁸⁾ and THz time-domain spectroscopy.^{9,10)}

Although the working principle of dual-comb spectroscopy is similar to that of conventional Fourier transform spectroscopy,¹¹⁾ dual-comb spectroscopy has several advantages: high data acquisition rate, high-frequency resolution, and no need for a mechanical delay stage. Dual-comb spectroscopy with two mode-locked lasers, whose repetition frequencies are given by f_{rep} and $f_{\text{rep}} + \Delta$, corresponds to Fourier transform spectroscopy with a scanning range of c/f_{rep} and an acquisition time of as short as $1/\Delta$, where f_{rep} , Δ , and c are the repetition frequency of one of the mode-locked lasers, the repetition frequency difference between two mode-locked lasers, and the velocity of light, respectively. Considering the spectral domain, dual-comb spectroscopy is equivalent to coherent multiheterodyne spectroscopy. Since the comb structure in the optical domain is down-converted into a radio frequency (RF) domain, long coherent averaging enables the obtained spectrum to be resolved in a number of comb lines. Resolved comb lines have 1 Hz linewidth with 1 s data acquisition, resulting in an ultimate frequency resolution of as high as 1 Hz when a stabilized frequency comb is employed.⁵⁾ However, tight phase locking between two mode-locked lasers is required in order to obtain accurate comb-to-comb heterodyning in dual-comb spectroscopy. Since mode-locked lasers have two degrees of freedom—repetition frequency and offset frequency—phase locking requires two references. A conventional method is to phase lock two mode-locked lasers at two different spectral components onto cavity-stabilized continuous-wave (CW) lasers.^{5,12)} Another method requires the post correction of data, using the phase information of beats between free-running CW lasers and mode-locked lasers.⁷⁾ Although the former has demonstrated outstanding

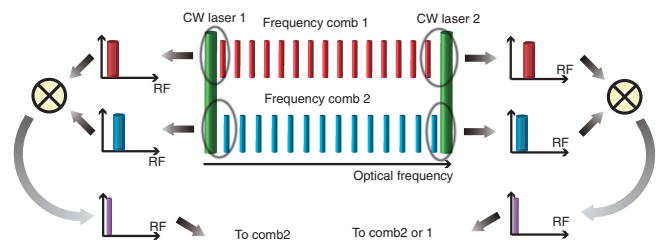


Fig. 1. Schematic of the simple phase-locking method for dual-comb spectroscopy. Two mode-locked lasers are relatively phase-locked by using two free-running CW lasers.

dual-comb spectroscopy with Hz-level accuracy, it requires expensive cavity-stabilized CW lasers with expertise in frequency metrology and laser engineering. Although the latter is cost-effective, it is hard to realize the absolute frequency measurement, and the data processing is somewhat complex. Very recently, adaptive dual-comb spectroscopy, which utilizes free-running CW lasers and devises the sampling clock, has also demonstrated comb-resolved dual-comb spectroscopy.^{13,14)}

In this letter, we propose a simple phase-locking method between two mode-locked lasers and demonstrate comb-resolved dual-comb spectroscopy. We achieved the transform-limited linewidth to the acquisition time, and found that this method could be used for the absolute frequency measurement.

Figure 1 shows the schematic of the phase-locking concept. Our proposed dual-comb spectroscopy is based on the relative phase locking between two frequency combs. Relative phase locking can be achieved by controlling repetition rate and offset frequency of comb2 to follow comb1, while comb1 is kept totally free running. Instead, it is also possible to control both comb1 and comb2 via, for example, pump LD current of both lasers. To realize relative phase locking, beats between comb1 and comb2 at two different spectral components need to be stabilized. Since the direct observation of RF beats between two combs is not possible because of low comb-mode power, free-running single-mode CW lasers are utilized as common frequency references.¹³⁾ The strong longitudinal-mode power of CW lasers makes it possible to observe beating against a weak comb mode with a good signal-to-noise ratio. By mixing the comb1-CW beat with the comb2-CW beat, the frequency noise of a CW laser is

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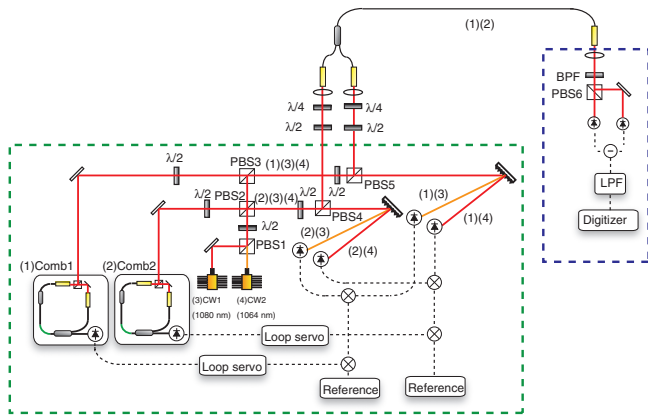


Fig. 2. Schematic of the experimental setup. PBS, polarized beam splitter; BPF, band-pass filter; LPF, low pass filter. The phase locking unit is shown inside the dotted green line. The dotted blue line shows the detection unit.

canceled very accurately. This is similar to the common-mode rejection in the differential amplifier in an RF circuit. The obtained differential signal corresponds to the beating between comb1 and comb2. The bandwidth of the common-mode rejection process is limited only by RF components (e.g., detectors, mixers, filters, and cables) and is expected to be as high as 400 MHz, limited by the detector in our case. Therefore, high stability and accuracy are not required for reference CW lasers, and a cost-effective, compact CW laser can be utilized. Finally, by stabilizing the obtained beatings between comb1 and comb2 at two different spectral components, two mode-locked lasers are relatively phase locked.

Figure 2 shows a schematic of the experimental setup. Comb1 and comb2 are Yb-doped fiber-based mode-locked lasers.¹⁵⁾ Their repetition frequencies are approximately 200 MHz. CW lasers are fiber Bragg-grating wavelength-stabilized single-longitudinal-mode diode lasers (QFBGLD-1060-10 and QFBGLD-1080-2 from QPhotonics, LLC). Their linewidths are specified to be below 1 MHz. A mode-locked Yb-doped fiber laser is superposed on CW lasers, and the overlapped beam is spectrally filtered by a grating and detected by avalanche photodiodes to observe beats between the mode-locked laser and the CW lasers at around 1064 and

1080 nm [Fig. 3(a)]. These beats have a large phase noise and a slow drift [Figs. 3(a) and 3(c)] mostly from CW lasers. Figure 3(b) shows the beating between comb1 and comb2, which can be obtained by mixing the beatings between CW lasers and combs. Because of the broadband common-mode rejection, the linewidth of the obtained beat frequency is as narrow as 100 kHz, limited by the resolution bandwidth of the RF spectrum analyzer, while the beat signal with CW lasers has a broader linewidth [Figs. 3(a) and 3(b)]. In addition, beatings between comb1 and comb2 drift only a few MHz in a few hundreds of seconds [Fig. 3(c)]. The obtained beatings between comb1 and comb2 at 1064 and 1080 nm are filtered out by RF circuits, compared with the RF reference by a digital phase detector, and feedback to the pump current of the oscillator. Although there are two feedback loops, the cross coupling did not disturb the phase locking so much. Figures 3(d) and 3(e) show phase-locked RF signals. Coherent carrier peaks with over 30 dB from the incoherent component were obtained at 1 kHz resolution bandwidth, confirming that approximately 80% of the total power is contained in the coherent carrier. If tighter phase locking is preferred, one of the error signals controls intracavity electro optic modulation,¹⁶⁾ or extracavity acousto optic modulation,¹²⁾ instead of pump current, to suppress the cross coupling between the feedback loops.

Although our CW lasers are observed to have a rather narrow linewidth (below 1 MHz), we expect that similar tight phase locking between two combs can also be performed with CW lasers with a much broader linewidth and a larger frequency noise because of the large common-mode rejection bandwidth. The average power of the CW lasers affects the signal-to-noise ratio of the beat signal. We found that the average power of the CW laser (0.1 mW) utilized in our demonstration is sufficient for obtaining a fine beatnote, as shown in Fig. 3. The slow drift between the CW laser and the frequency combs must be suppressed below half the repetition frequency over the measurement time for the application where long-term phase locking is desirable. This can be done passively by improving the temperature stability around the CW lasers or actively by controlling the pump current of the CW lasers.

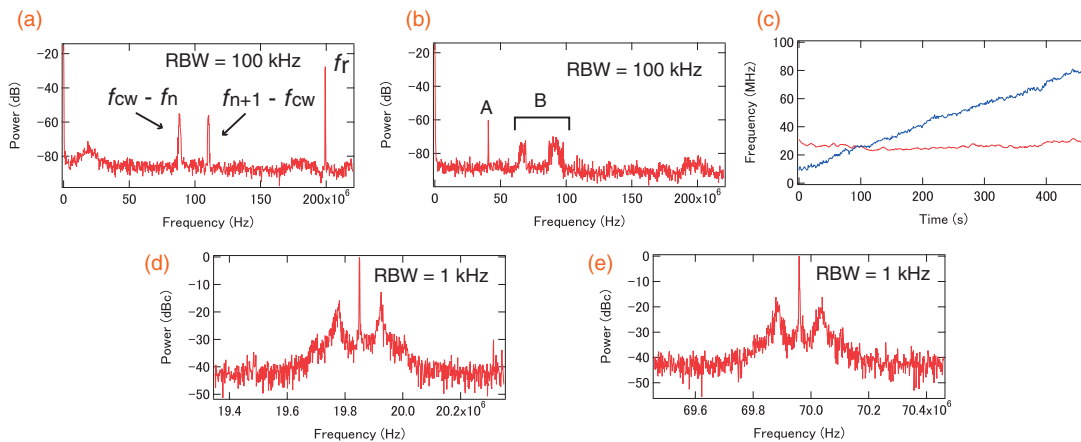


Fig. 3. (a) RF spectrum of beatnote between a mode-locked laser and a free-running cw laser. (b) Beatnote between two mode-locked lasers obtained after mixing two RF beats: CW-comb1 and CW-comb2 [A]. Peaks in region B are RF beats that originate from other (unwanted) combinations between the CW and mode-locked lasers. (c) (blue) Drift of the rf beat between comb and cw laser. (red) Drift of the rf beat between comb1 and comb2. Locked beat signals at (d) 1064 nm and (e) 1080 nm.

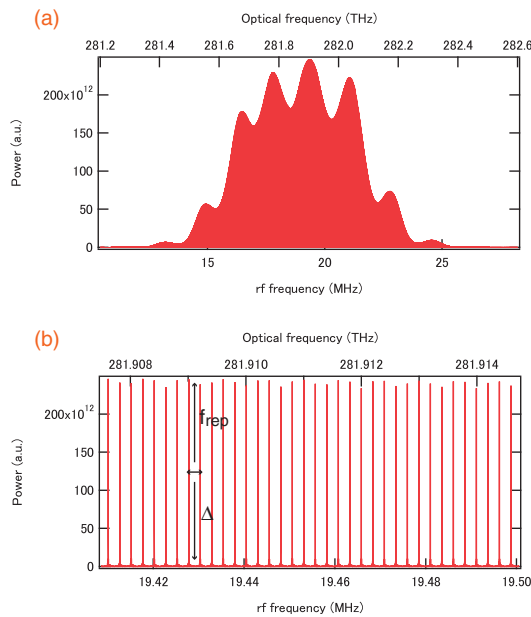


Fig. 4. (a) RF spectrum generated from the Fourier transform of time-domain interferograms at 150 ms. (b) Expanded spectrum of part (a).

To demonstrate dual-comb spectroscopy with the above-mentioned relatively locked frequency combs, the repetition frequency difference between those combs is set to approximately 2.5 kHz. Comb1 from PBS5 and comb2 from PBS4 in Fig. 2 are spatially overlapped by utilizing a fiber coupler. Wave plates before the fiber coupler are used for adjusting the power ratio between horizontal and longitudinal polarizations. Band-pass filter at the center wavelength of 1064 nm with 3 nm bandwidth is inserted before the PBS6 in order not only to limit the optical bandwidth, but also to work as a sample. The DC component is eliminated by applying balanced detection. The detected interferometric signal between the two mode-locked lasers is digitized at a 200 MHz sampling rate by a 14-bit analog-to-digital converter. Before digitizing, a 100 MHz low-pass filter smoothens the pulsed signal to obtain a reliable temporal overlap between pulse arrival and sampling times. To utilize the full range of the AD converter, the signal amplitude is optimized by an RF amplifier before digitizing. The 14-bit AD converter has a 2 GB of memory and is capable of employing an external clock up to 200 MHz. Currently, our maximum measurement time is limited by the available memory of the AD converter. An AD converter with more working memory has been developed and would enable higher-resolution dual-comb spectroscopy with a longer measurement time. By Fourier transforming the time-domain interferograms taken at a 150 ms acquisition time, finely resolved comb modes are observed a bandwidth of 1 THz limited by the optical band-pass filter [Fig. 4(a)]. In the RF domain, the spectrum consists of numerous longitudinal modes with the mode spacing given by the repetition frequency difference [Fig. 4(b)]. The spectrum can also be shown in optical frequency by calibrating the RF frequency to optical frequency as shown in the top vertical axis of Fig. 4(b). The spectrum has a mode spacing of the repetition frequency. Resolved comb modes with a transform-limited linewidth of approximately 6 Hz in the RF domain are obtained. The coherence between two combs in

different frequency regions is estimated by evaluating the linewidth of the RF combs. It is found that the linewidths in three different spectral regions across an optical bandwidth of approximately 1 THz are inversely proportional to the data acquisition time. The result confirms that tight phase locking between two mode-locked lasers is sustained for at least 150 ms to offer a Fourier-limited linewidth for the beatnotes. Since the bandwidth of dual-comb spectroscopy is limited to $f_{\text{rep}}^2/2\Delta$ to avoid aliasing, a smaller repetition frequency difference expands the bandwidth as long as two mode-locked lasers are coherent over the entire bandwidth.

Although dual-comb spectroscopy stabilized by frequency-calibrated CW lasers^{5,12)} naturally gives an absolute frequency calibration, our demonstrated dual-comb spectroscopy only offers spectra with an unknown frequency offset. This is because the free-running mode-locked lasers have an inevitable drift in those frequency modes even after relative phase locking. We experimentally determined that two frequency combs drift by approximately 1 MHz in optical frequency within 1 s. When an absolute frequency-calibrated spectrum is required with our proposed simple phase-locking method, one of the frequency combs can be frequency stabilized. We confirmed that dual-comb spectroscopy with a performance similar to that indicated in the previous section can also be performed when the relative phase locking between two combs is achieved by controlling the pump current and intracavity piezoelectric transducer of comb2, while comb1 is kept totally free-running. Thus, by simply stabilizing the repetition and offset frequencies of comb1 as is conventionally done,¹⁷⁾ dual-comb spectroscopy with absolute frequency calibration can be expected.

In conclusion, we have demonstrated a simplified phase-locking method between two mode-locked lasers for dual-comb spectroscopy. Two mode-locked lasers are relatively stabilized by utilizing CW lasers as the common frequency reference. Since the frequency noise of the CW lasers is canceled by the common-mode rejection, free-running CW lasers can be used for that purpose. In this way, comb-resolved dual-comb spectroscopy is demonstrated with 1 THz bandwidth. Our simple phase-locking method is expected to expand the use of dual-comb spectroscopy.

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