Dual-comb spectroscopy with a phase-modulated probe comb for sub-MHz spectral sampling

NICOLAS BOURBEAU HÉBERT[1], VINCENT MICHAUD-BELLEAU[1], SÉBASTIEN MAGNAN-SAUCIER[1], JEAN-DANIEL DESCHÊNES[1], AND JÉRÔME GENEST[1,*]

[1] Centre d'optique, photonique et laser, Université Laval, Québec, QC, G1V 0A6, Canada
[*] Corresponding author: jgenest@gel.ulaval.ca

Compiled July 25, 2016

We present a straightforward and efficient method to reduce the mode spacing of a frequency comb based on binary pseudo-random phase modulation of its pulse train. As a proof of concept, we use such a densified comb to perform dual-comb spectroscopy of a long-delay Mach-Zehnder interferometer and of a high-quality-factor microresonator with sub-MHz spectral sampling. Since this approach is based on binary phase modulation, it combines all the advantages of other densification techniques: simplicity, single-step implementation, and conservation of the initial comb's power.

OCIS codes: *(120.5060) Phase modulation; (140.4050) Mode-locked lasers; (300.6310) Spectroscopy, heterodyne; (300.6320) Spectroscopy, high-resolution; (300.6380) Spectroscopy, modulation.*

1. INTRODUCTION

Precision spectroscopy has greatly benefited from the the advent of mode-locked frequency combs. These broadband light sources generate well known and evenly spaced spectral modes that can be used to measure optical transfer functions (OTF) on an absolute frequency scale [1, 2]. However, the density of modes is difficult to modify as it is fixed by the comb's cavity length. Comb mode spacings ranging from a few tens of MHz to several GHz are easily accessible with typical mode-locked combs, which are well suited for Doppler- and pressure-broadened molecular spectroscopy [3]. On the other hand, atomic and Doppler-free spectroscopy as well as the measurement of high-quality-factor resonators [4] generally require denser combs to appropriately sample a typical OTF.

Several methods can be used to obtain denser spectral sampling in frequency comb spectroscopy. For instance, changing the repetition rate or carrier-enveloppe offset frequency moves the comb modes and allows the reconstruction of a spectrum with higher density by interleaving multiple measurements [5–8]. This approach works, but can be cumbersome to use in practice as it typically implies a re-tuning of the comb's parameters between each acquisition. The mode spacing can also be externally reduced by gating pulses using an intensity modulator [9, 10], but this is necessarily associated with an intrinsic power loss proportional to the pulse gating ratio. Alternatively, multilevel phase modulation has been proposed to achieve the same goal through the Talbot effect [11], but this approach requires a fast and expensive arbitrary waveform generator. Even though special cases of this approach can work with binary phase modulation, they are limited to a densification factor of 4 [12]. Densification techniques are not only of interest for spectroscopy, but also find applications in the stabilisation of large-mode-spacing combs [13], in the synthesis of stable microwave signals [14], and in unambiguous laser ranging [9].

In this Letter, we demonstrate a new approach to reduce the mode spacing of a frequency comb for spectroscopic applications. We show that phase modulating its pulse train with a pseudo-random binary sequence (PRBS) generates new comb modes with a spacing equivalent to the repetition rate of the binary sequence. Any densification factor of the form $2^m - 1$, with m an integer, is easily implemented. This technique allows to obtain dense spectra in a single experimental step and it combines the simplicity of pulse gating, which only requires a digital pattern generator to drive the modulator, with the power efficiency of phase modulation. Indeed, our approach is lossless because it uses phase rather than intensity modulation, and allows an equal redistribution of the power amongst the newly generated modes. We demonstrate its capabilities by densifying a 100-MHz mode-locked frequency comb by factors of 7, 31 and 127. We then use the resulting combs to perform quasi-integer-ratio dual-comb spectroscopy [15] and test our approach by probing a Mach-Zehnder interferometer having a 40-MHz free spectral range (FSR). We finally measure the complex OTF of a microresonator that shows resonances as narrow as 15 MHz with a spectral point spacing of 787 kHz.

2. METHODS

Figure 1 sketches the experimental setup used for this demonstration. A 100-MHz probe comb (Menlo Systems) is sent to an electro-optic phase modulator (EOM), which is driven by a maximum-length PRBS [16] of length $N = 2^m - 1$ generated by a digital pattern generator (DPG) (Anritsu ME522A). The bit rate of the sequence is synchronised and phase matched to the probe comb so that bit transitions occur between optical pulses. Its amplitude is in turn set close to the half-wave voltage of the modulator (V_π). Thus, optical pulses see 0 and π phase shifts following the binary sequence. The result is a densified comb with a new mode spacing of $100/N$ MHz, where modes have a pseudo-random phase relationship. This comb is used to interrogate a device under test (DUT) before being combined with a slightly detuned local oscillator (LO) comb running at 100 MHz $+\Delta f$. This setup is that of a dual-comb spectrometer [3] operating in a quasi-integer-ratio mode [15]. The beat note is measured with a balanced photodetector (Thorlabs PDB130C) and a digitiser (ADC) (14 bits, 100 MHz), which is synchronised with the LO comb (100 MS/s) so that a single electrical sample is recorded at the arrival of each LO pulse. A referencing stage tracks the relative drifts of the combs, which are later accounted for in a post-correction step [17]. This allows us to resolve the densified comb regardless of the relative drifts.

Not shown in Fig. 1 are two optical filters (JDSU TB9) with full width at half maximum of 30 GHz, one for each comb, and a calibration channel measuring the mixing product between the LO comb and the densified probe comb before the DUT. The necessity of the filters is discussed in section 4.

In contrast to electro-optic combs generated by pseudo-random modulation of a continuous-wave laser [18], here the bandwidth is defined by the original comb rather than by the modulation bandwidth because the pattern is sampled by much shorter pulses. One thus only needs to generate a PRBS with bandwidth greater than the comb's repetition rate. That is, the DPG sets the limit on the fastest comb that can be densified.

Figure 2 illustrates the working principle in the time and frequency domains. In Fig. 2(a), a probe pulse train (black) is depicted after phase modulation by a $N = 3$ sequence. A LO pulse train (multicoloured) with repetition period T_{LO} slightly detuned from that of the probe pulse train is also shown. Figure 2(b) shows the response (black) of an arbitrary DUT to the probe pulse train. The corresponding impulse response is shown with an offset (dashed black). As the impulse response is longer than the delay between two probe pulses, the resulting signal is the sum of many successive and overlapping responses. This means that the comb's initial spacing is too large to resolve the OTF. However, since the probe pulse train is encoded with pseudo-random phases, the effective probing period is N times longer than expected, which is sufficient to fully measure the impulse response shown here. The true impulse response can be recovered by cross-correlation with the PRBS or by using a calibration channel. Multicoloured points in Fig. 2(b) correspond to optical samples taken by the LO comb. As there are N samples taken per effective probing period, N independent and time-multiplexed interferograms (IGM) corresponding to the time-stretched response of the DUT to the probing sequence, as the one shown by the blue dashed curve, are constructed simultaneously [15].

Figure 2(c) shows the LO comb (multicoloured) in the optical frequency domain along with the probe comb (black) densified using pseudo-random phase modulation with $N = 3$. The newly generated modes are easily made spectrally flat using a maximum-length PRBS, while the relative intensity of the original modes and the new modes can be adjusted with the amplitude of the binary sequence. The raw radio frequency (RF) spectrum resulting from the beating between the LO and probe combs is given in Fig. 2(d). We notice the emergence of several groups of modes containing information about different optical frequencies. RF modes can be associated to beating pairs of probe and LO modes from Fig. 2(c) using the numbers and the colour code. Unnumbered modes are part of negative aliases of the spectrum and contain redundant information.

The raw data is processed as follows to produce a calibrated OTF. The signal is first demultiplexed by keeping one out of every N samples to yield N IGMs. The referencing signals are demultiplexed in the same way and are used to correct each IGM as in [17]. As long as the acquisition time is long enough, each of these N data sets is a full IGM and can simply be Fourier transformed to yield the DUT's transmittance. We instead coherently average them (after alignment by correlation) before taking the Fourier transform in order to recover the signal-to-noise ratio offered by the measurement. A spectral division by the calibration data, which is processed identically, is finally performed.

3. RESULTS

We first measure the OTF of a 5-meter-delay Mach-Zehnder (MZ) fibre interferometer (FSR = 40 MHz) with various mode spacings to validate our technique. We choose this device as a test bench because it displays a simple sine-shaped OTF . Figure 3 shows the device's OTF measured with 4 different mode spacings: (a) 100 MHz (no modulation), (b)

100/7 = 14.3 MHz, (c) 100/31 = 3.23 MHz and (d) 100/127 = 0.787 MHz. Points correspond to samples taken on the OTF by the probe comb's modes while solid lines of matching colours are obtained by ideal (sinc) interpolation. All measurements are calibrated using data provided simultaneously by a calibration channel.

In Fig. 3(a), the unmodulated probe comb does not have a sufficiently low mode spacing to properly sample the OTF of interest. Indeed, in analogy to time sampling, frequency sampling is unambiguous if there are at least two samples per shortest period of the spectral signal. In the present case, the shortest period is 40 MHz and aliasing occurs since the spectral point spacing is superior to 40/2 = 20 MHz. With all three scenarios depicted in Fig. 3(b), (c) and (d), the mode spacing is low enough so that the true OTF is measured. It is important to note that as the mode spacing is decreased, the comb's power is redistributed amongst more modes while the noise power stays at a constant level. This results in a signal-to-noise ratio that is reduced by a factor N, which is the minimum penalty associated with a densification of the comb by the same factor.

We also use this technique to measure the complex OTF of a spherical silica microresonator (diameter of 300 µm) evanescently coupled by a tapered fibre [19], which is of greater experimental interest. Such resonators show very sharp resonances and thus require dense spectral sampling. We use a PRBS with $N = 127$, resulting in a spectral point spacing of 787 kHz. Figure 4(a) shows the temporal response of the microresonator to a complete probing sequence whose duration is the inverse of the modified spectral point spacing. The sequence contains $N = 127$ pulses with pseudo-randomly alternating phase shifts. The inset shows the ring-down signal after a pulse.

The device's spectral transmittance around 1562.23 nm is given in Fig. 4(b) over a 40-GHz range, which is limited by the optical filters mentioned earlier. Several resonances with widths as small as 15 MHz can be observed, justifying the need for densification. Some of them also appear to be slightly asymmetrical. We believe this is due to a Fano effect caused by a slightly multimode taper since our experimental conditions are very similar to those in [20]. The corresponding phase spectrum is shown in Fig. 4(c). Two kinds of resonances with excursions of either 2π or in the 0-π range can clearly be seen. This indicates the presence of both overcoupled and undercoupled resonances [21], respectively. The measurement time is $\sim$ 2.5 s, which corresponds to the acquisition card's maximum capacity, and the number of resolved spectral elements is $\sim$ 50000. This gives rise to a peak signal-to-noise ratio before calibration with the calibration channel of $\sim$ 40 dB. We calculate it as the ratio of the spectrum's maximum value to the out-of-band noise floor.

4. DISCUSSION

In the present demonstration, both combs are free-running and their relative drifts are post-corrected using signals provided by the referencing stage. Although drifts coming from the combs themselves can be accounted for, residual drifts caused by components and fibres that are part of the dual-comb interferometer cannot be compensated by the referencing stage (see Fig. 1). The same issue would occur with actively stabilised combs since the signals used for stabilisation and the signal used for the measurement of interest cannot travel through identical paths. To ensure that negligible drifts occur within a single IGM, a high IGM scanning rate, governed by the repetition rate detuning (Δf) between the combs, must be used. As the mode spacing is reduced, the IGM length grows and the scanning rate must be increased accordingly to maintain a constant fibre noise immunity. This however results in a RF spectrum with increased bandwidth and thus optical filtering becomes necessary at some point to prevent the RF spectrum from exceeding the comb's Nyquist frequency. For this proof of concept, we filter both combs with identical 30-GHz filters and use $\Delta f = 2$ kHz, 500 Hz and 125 Hz for the cases $N = 7$, 31 and 127, respectively. This ensures that, in all cases, the IGMs are scanned quickly enough without violating the Nyquist criterion and that approximately the same number of IGMs is generated for coherent averaging. Reduction of the fibre noise with a vibration-immune and thermally-stable setup or with active stabilisation would allow to access the full comb's bandwidth through the use of a lower Δf.

The spectral resolution in comb-based spectroscopy is always ultimately limited by the linewidth of the probe comb during the measurement time. Here, the linewidth is that of the initial free-running comb ($<$ 1 MHz at 2.5 s). Since the spacing between the modes is usually much larger than their width, spectral resolution and spectral point spacing are two distinct quantities. The goal of this densification technique is thus to bridge the gaps between modes and reach the limit where the two quantities are equal. This situation corresponds to the maximum useful densification. Although we established the viability of this technique for dual-comb spectroscopy, further work will be necessary to explore the resolution limit imposed by the linewidth. Ultimately, active stabilisation of the probe comb could afford the highest resolution and be a good match for this densification technique.

Finally, it is worth mentioning that spectra obtained with this technique are likely to show periodic errors if the measurement and calibration channels do not have identical transfer functions. In a conventional dual-comb setup, a mismatch between the detectors translates into a slowly varying error on the calibrated RF spectrum. However, the same mismatch translates into a periodic error (100 MHz in this case) with the approach presented here since the RF spectrum actually consists of distant groups of modes that are to be interleaved (see Fig. 2(d)). A correction of this effect relying on the characterisation of our detectors' mismatch is included in the measurements presented above.

5. CONCLUSION

We showed that the mode spacing of a frequency comb can be efficiently reduced by phase modulating its pulse train with a pseudo-random binary sequence. This simple approach is of great interest for spectroscopy as it provides tunability of the spectral sampling density independently of the repetition rate, which is rather constrained by laser optimisation considerations. The only hardware that needs to be added to a conventional dual-comb setup is a DPG and an EOM. We validated our approach by measuring the sinusoidal OTF of a Mach-Zehnder interferometer with various spectral point spacings. We also measured the complex OTF of a microresonator with a spectral point spacing of 787 kHz, which corresponds to a 127-fold increase in spectral density. This is comparable to the spectral density obtained in [6], but our approach achieved this with a single continuous acquisition. The densification technique presented here could be combined with other types of comb-based spectrometers, such as those relying on spatial dispersion, and should thus be useful to a vast portion of the spectroscopy community.

This research was supported by the Natural Sciences and Engineering Research Council of Canada (NSERC) and the Fonds de recherche du Québec - Nature et technologies (FRQNT).

REFERENCES

1. R. Holzwarth, T. Udem, T. W. Hänsch, J. Knight, W. Wadsworth, and P. S. J. Russell, Phys. Rev. Lett. **85**, 2264 (2000).
2. A. Marian, M. C. Stowe, D. Felinto, and J. Ye, Phys. Rev. Lett. **95**, 023001 (2005).
3. I. Coddington, W. C. Swann, and N. R. Newbury, Phys. Rev. Lett. **100**, 013902 (2008).
4. G. Lin, S. Diallo, R. Henriet, M. Jacquot, and Y. K. Chembo, Opt. Lett. **39**, 6009 (2014).
5. Y.-D. Hsieh, Y. Iyonaga, Y. Sakaguchi, S. Yokoyama, H. Inaba, K. Minoshima, F. Hindle, T. Araki, and T. Yasui, Sci. Rep. **4** (2014).
6. S. Okubo, Y.-D. Hsieh, H. Inaba, A. Onae, M. Hashimoto, and T. Yasui, Opt. Express **23**, 33184 (2015).
7. S. A. Diddams, L. Hollberg, and V. Mbele, Nature **445**, 627 (2007).
8. J. D. Anstie, C. Perrella, P. S. Light, and A. N. Luiten, Opt. Express **24**, 4088 (2016).
9. S. Boudreau, S. Levasseur, C. Perilla, S. Roy, and J. Genest, Opt. Express **21**, 7411 (2013).
10. V. Supradeepa, D. E. Leaird, and A. M. Weiner, Opt. Express **17**, 14434 (2009).
11. A. Malacarne and J. Azaña, Opt. Express **21**, 4139 (2013).
12. M. Beltran, J. Caraquitena, R. Llorente, and J. Marti, IEEE Photon. Technol. Lett. **16**, 1175 (2011).
13. P. Del'Haye, S. B. Papp, and S. A. Diddams, Phys. Rev. Lett. **109**, 263901 (2012).
14. J. Li, X. Yi, H. Lee, S. A. Diddams, and K. J. Vahala, Science **345**, 309 (2014).
15. N. B. Hébert, S. Boudreau, J. Genest, and J.-D. Deschênes, Opt. Express **22**, 29152 (2014).
16. S. Haykin, *Communication systems* (John Wiley & Sons, 2001).
17. J.-D. Deschênes, P. Giaccari, and J. Genest, Opt. Express **18**, 23358 (2010).
18. N. B. Hébert, V. Michaud-Belleau, J. D. Anstie, J.-D. Deschênes, A. N. Luiten, and J. Genest, Opt. Express **23**, 27806 (2015).
19. V. Michaud-Belleau, J. Roy, S. Potvin, J.-R. Carrier, L.-S. Verret, M. Charlebois, J. Genest, and C. N. Allen, Opt. Express **20**, 3066 (2012).
20. A. Chiba, H. Fujiwara, J.-i. Hotta, S. Takeuchi, and K. Sasaki, Appl. Phys. Lett. **86**, 261106 (2005).
21. A. B. Matsko, *Practical applications of microresonators in optics and photonics* (CRC Press, 2009).

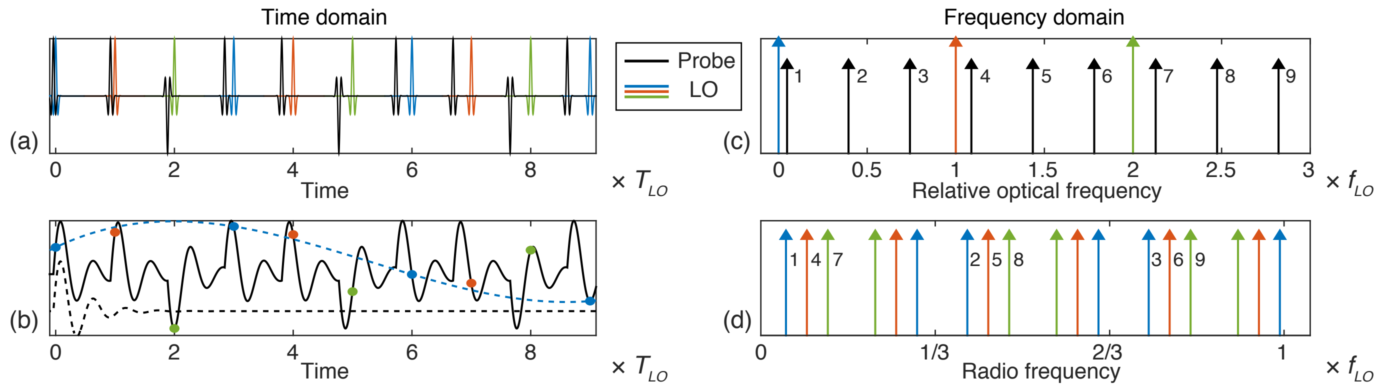

Fig. 2. Dual-comb spectroscopy using a comb densified with pseudo-random phase modulation. (a) Probe (black) and LO (multicoloured) pulse trains. The probe is modulated using a $N = 3$ sequence. (b) Response of an arbitrary DUT to the probe pulse train (black) and optical samples (multicoloured) taken by the LO pulse train. The 3 subsets of LO pulses (blue, orange and green) each generate an independent IGM (as the one shown by the blue dashed curve). (c) Densified probe comb (black) and LO comb (multicoloured). (d) RF spectrum generated from the beating of the probe and LO combs.

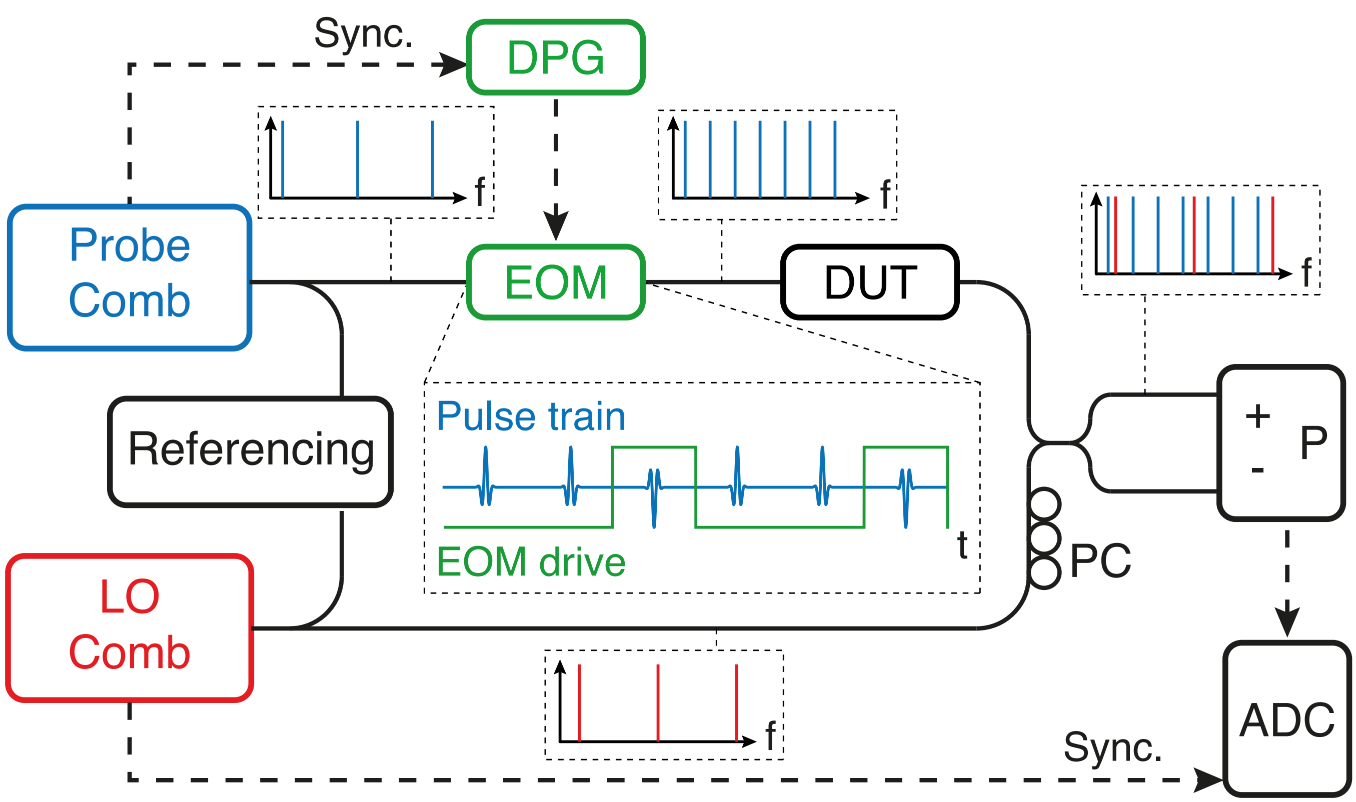

Fig. 1. Experimental setup. The mode spacing of a probe comb is reduced using pseudo-random phase modulation. The resulting densified comb interrogates a DUT and is combined with a LO comb to perform dual-comb spectroscopy. The modulation sequence shown here has a length of $N = 3$. PC: polarisation controller. P: balanced photodetector.

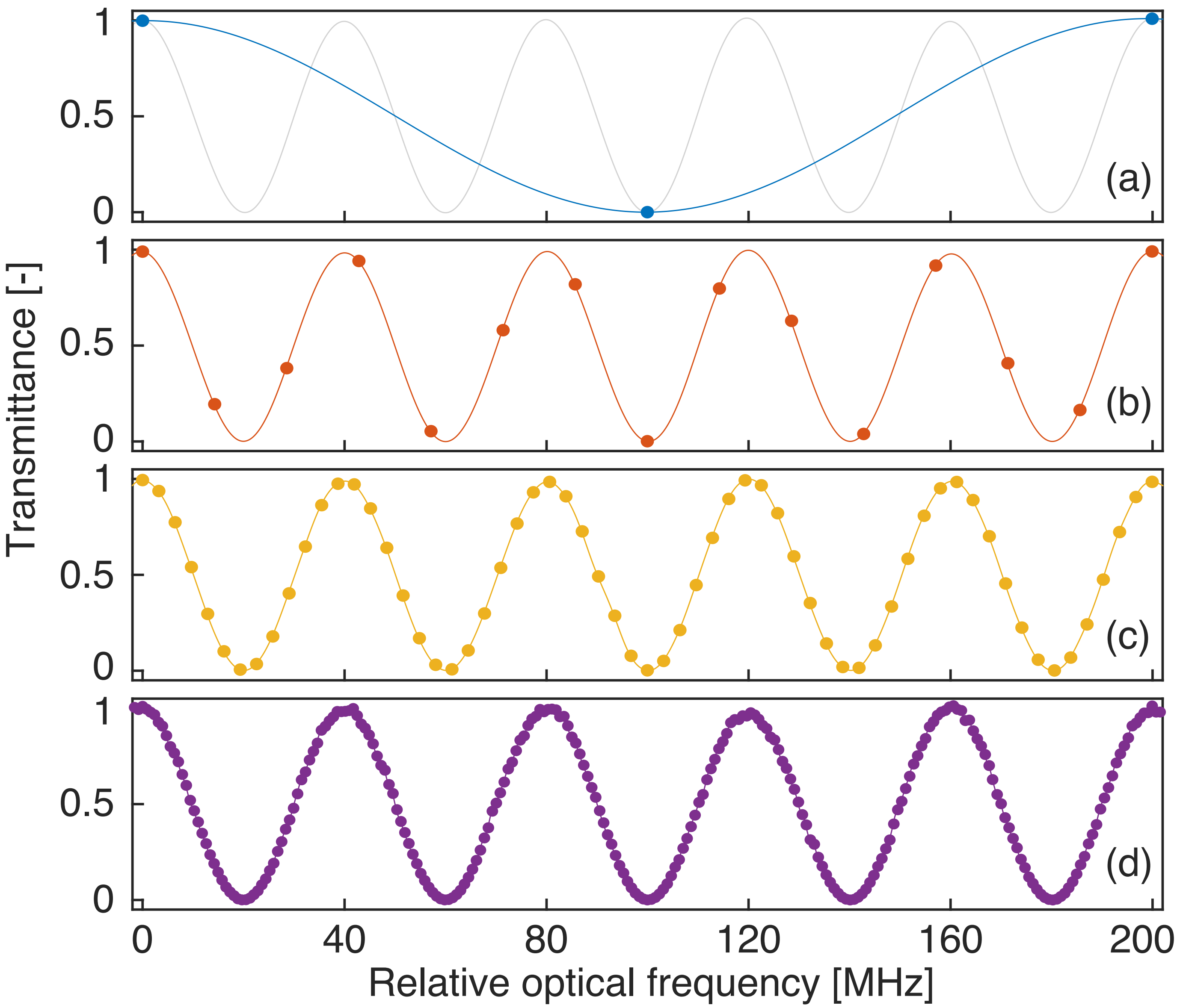

Fig. 3. Spectral transmittance of a 5-m-delay Mach-Zehnder interferometer (FSR = 40 MHz). (a) OTF probed with the unmodulated comb. The spectral point spacing is 100 MHz and the FSR falsely appears to be 200 MHz because of aliasing. The same OTF is probed using different sequence lengths to obtain measurements with spectral point spacings of (b) $100/7 = 14.3$ MHz, (c) $100/31 = 3.23$ MHz and (d) $100/127 = 0.787$ MHz.

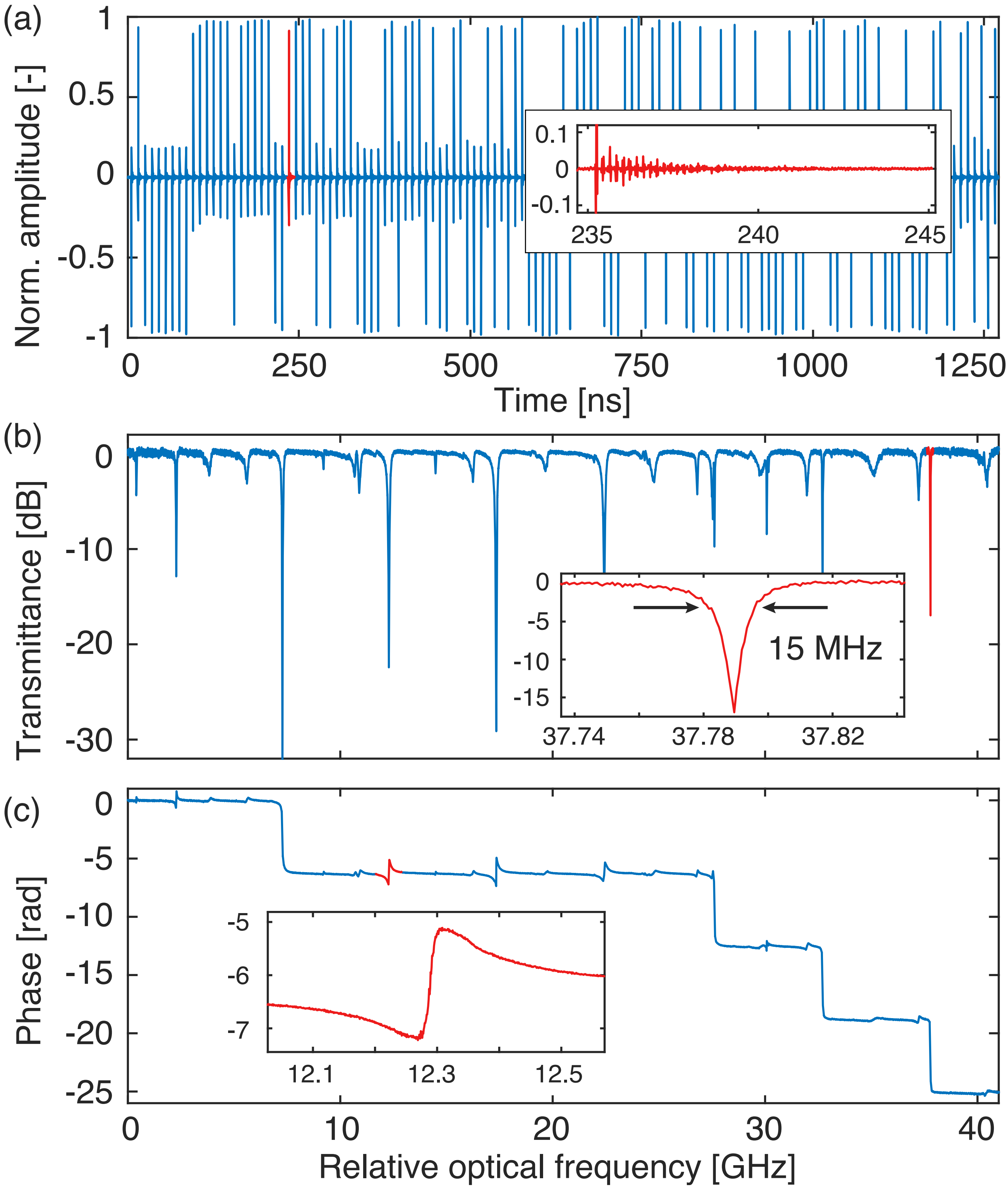

Fig. 4. (a) Temporal response of the microresonator to a $N = 127$ pseudo-random sequence of pulses each spaced by 10 ns. (b) Calibrated transmittance of the microresonator with 787-kHz spectral point spacing. The inset shows a resonance that has a 3-dB width of 15 MHz. The frequency axis is relative to one of the continuous-wave lasers used in the referencing stage ($\sim$ 1562.23 nm). (c) Corresponding phase spectrum showing both overcoupled and undercoupled resonances.

REFERENCES

1. R. Holzwarth, T. Udem, T. W. Hänsch, J. Knight, W. Wadsworth, and P. S. J. Russell, “Optical frequency synthesizer for precision spectroscopy,” Phys. Rev. Lett. **85**, 2264 (2000).
2. A. Marian, M. C. Stowe, D. Felinto, and J. Ye, “Direct frequency comb measurements of absolute optical frequencies and population transfer dynamics,” Phys. Rev. Lett. **95**, 023001 (2005).
3. I. Coddington, W. C. Swann, and N. R. Newbury, “Coherent multiheterodyne spectroscopy using stabilized optical frequency combs,” Phys. Rev. Lett. **100**, 013902 (2008).
4. G. Lin, S. Diallo, R. Henriet, M. Jacquot, and Y. K. Chembo, “Barium fluoride whispering-gallery-mode disk-resonator with one billion quality-factor,” Opt. Lett. **39**, 6009–6012 (2014).
5. Y.-D. Hsieh, Y. Iyonaga, Y. Sakaguchi, S. Yokoyama, H. Inaba, K. Minoshima, F. Hindle, T. Araki, and T. Yasui, “Spectrally interleaved, comb-mode-resolved spectroscopy using swept dual terahertz combs,” Sci. Rep. **4** (2014).
6. S. Okubo, Y.-D. Hsieh, H. Inaba, A. Onae, M. Hashimoto, and T. Yasui, “Near-infrared broadband dual-frequency-comb spectroscopy with a resolution beyond the fourier limit determined by the observation time window,” Opt. Express **23**, 33184–33193 (2015).
7. S. A. Diddams, L. Hollberg, and V. Mbele, “Molecular fingerprinting with the resolved modes of a femtosecond laser frequency comb,” Nature **445**, 627–630 (2007).
8. J. D. Anstie, C. Perrella, P. S. Light, and A. N. Luiten, “Coherent radio-frequency detection for narrowband direct comb spectroscopy,” Opt. Express **24**, 4088–4096 (2016).
9. S. Boudreau, S. Levasseur, C. Perilla, S. Roy, and J. Genest, “Chemical detection with hyperspectral lidar using dual frequency combs,” Opt. Express **21**, 7411–7418 (2013).
10. V. Supradeepa, D. E. Leaird, and A. M. Weiner, “Single shot amplitude and phase characterization of optical arbitrary waveforms,” Opt. Express **17**, 14434–14443 (2009).
11. A. Malacarne and J. Azaña, “Discretely tunable comb spacing of a frequency comb by multilevel phase modulation of a periodic pulse train,” Opt. Express **21**, 4139–4144 (2013).
12. M. Beltran, J. Caraquitena, R. Llorente, and J. Marti, “Reconfigurable multiwavelength source based on electrooptic phase modulation of a pulsed laser,” IEEE Photon. Technol. Lett. **16**, 1175–1177 (2011).
13. P. Del’Haye, S. B. Papp, and S. A. Diddams, “Hybrid electro-optically modulated microcombs,” Phys. Rev. Lett. **109**, 263901 (2012).
14. J. Li, X. Yi, H. Lee, S. A. Diddams, and K. J. Vahala, “Electro-optical frequency division and stable microwave synthesis,” Science **345**, 309–313 (2014).
15. N. B. Hébert, S. Boudreau, J. Genest, and J.-D. Deschênes, “Coherent dual-comb interferometry with quasi-integer-ratio repetition rates,” Opt. Express **22**, 29152–29160 (2014).
16. S. Haykin, *Communication systems* (John Wiley & Sons, 2001).
17. J.-D. Deschênes, P. Giaccari, and J. Genest, “Optical referencing technique with cw lasers as intermediate oscillators for continuous full delay range frequency comb interferometry,” Opt. Express **18**, 23358–23370 (2010).
18. N. B. Hébert, V. Michaud-Belleau, J. D. Anstie, J.-D. Deschênes, A. N. Luiten, and J. Genest, “Self-heterodyne interference spectroscopy using a comb generated by pseudo-random modulation,” Opt. Express **23**, 27806–27818 (2015).
19. V. Michaud-Belleau, J. Roy, S. Potvin, J.-R. Carrier, L.-S. Verret, M. Charlebois, J. Genest, and C. N. Allen, “Whispering gallery mode sensing with a dual frequency comb probe,” Opt. Express **20**, 3066–3075 (2012).
20. A. Chiba, H. Fujiwara, J.-i. Hotta, S. Takeuchi, and K. Sasaki, “Fano resonance in a multimode tapered fiber coupled with a microspherical cavity,” Appl. Phys. Lett. **86**, 261106 (2005).
21. A. B. Matsko, *Practical applications of microresonators in optics and photonics* (CRC Press, 2009).