Widely tunable cavity-enhanced frequency combs

MYLES C. SIFLIES¹, GRZEGORZ KOWZAN², YUNING CHEN¹, NEOMI LEWIS¹, RYAN HOU³, ROBIN BAEHRE⁴, TOBIAS GROSS⁴, AND THOMAS K. ALLISON¹*

¹Departments of Physics and Chemistry, Stony Brook University, Stony Brook NY, 11794
²Institute of Physics, Faculty of Physics, Astronomy and Informatics, Nicolaus Copernicus University, ul. Grudziądzka 5/7, 87-100 Torun, Poland
³Department of Physics, Columbia University, New York NY, 10027
⁴Laseroptik GmbH, Horster Str. 20, 30826 Garbsen, Germany
*Corresponding author: thomas.allison@stonybrook.edu

We describe the cavity-enhancement of frequency combs over a wide tuning range of 450-700 nm (> 7900 cm⁻¹), covering nearly the entire visible spectrum. Tunable visible frequency combs from a synchronously-pumped optical parametric oscillator are coupled into a 4-mirror, dispersion-managed cavity with a finesse of 600 to 1400. An intracavity absorption path length enhancement greater than 190 is obtained over the entire tuning range, while preserving intracavity spectral bandwidths capable of supporting sub-200 fs pulse durations. These tunable cavity-enhanced frequency combs can find many applications in nonlinear optics and spectroscopy. © 2020 Optical Society of America

http://dx.doi.org/10.1364/ao.XX.XXXXXX

The cavity enhancement of stabilized ultrafast pulse trains, or frequency combs, first demonstrated around the turn of the century [1, 2], has since been used in many applications. In this technique, successive pulses from the frequency comb are constructively interfered with a circulating intracavity pulse by tuning both the comb’s repetition rate (f_rep) and carrier-envelope offset frequency (f₀) such that the frequency comb’s “teeth” are matched with the enhancement cavity resonance frequencies over a large spectral bandwidth. In some applications, this method is used to enhance the intracavity power to drive nonlinear processes at high repetition rate. For example, with kilowatts of circulating average power, one can generate high-order harmonics at high repetition rate [3], and this is now being used for precision spectroscopy [4] and high-repetition rate photoelectron spectroscopy experiments [5–7]. Other ultrafast nonlinear processes have also used the high intracavity power, such as molecular alignment [8] and spontaneous parametric down conversion [9]. In another class of applications, it is the enhancement of sensitivity that is sought, such as in cavity-enhanced direct frequency comb spectroscopy, reviewed by Adler et al. in 2010 [10] and continuing to make rapid progress since then [11–13]. A new application of cavity-enhanced frequency combs developed by our group uses the enhancement of both laser power and sensitivity to obtain a large improvement in the detection-limits of ultrafast nonlinear spectroscopy [14, 15].

In all of this previous work, the cavities have been carefully designed to enhance frequency combs with a certain center wavelength for a specifically targeted intracavity experiment, with limited bandwidth and little or no tuning range. In contrast, frequency comb experiments without cavities are increasingly using combs covering very wide spectral ranges, and the development of widely tunable and broadband frequency combs has recently been the subject of intense research [16–24].

The cavity-enhancement of frequency combs over a wide spectral range poses a number of technical challenges. First, one must have a tunable frequency comb with transducers to control both the f_rep and f₀ of the comb, with at least one high-bandwidth transducer to tightly lock the comb to the cavity or vice versa. This rules out, for example, the recently popular offset-free “DFG combs” [25, 26]. Second, the combs should have low optical phase noise. For typical repetition rates of ~ 100 MHz, even modest cavity fineses of ~ 1000 have small optical linewidths of ~ 100 kHz. The linewidths of the comb’s “teeth” should then be substantially narrower than this in order to avoid increased intensity noise on the intracavity light or a reduction in effective input power, and accomplishing this requires special care [27]. Third, in order to couple a large comb bandwidth, and thus short pulses, into the cavity, very good control of the cavity’s intracavity group-delay dispersion (GDD) must be achieved. For example, if a 100 fs input pulse duration is to remain less than 200 fs in a cavity with a finesse of 1000, the net GDD must be less than ~ 100 fs². Achieving intracavity GDD this low at one design wavelength is straightforward, but managing dispersion at this level over a wide tuning range, while also satisfying the constraints imposed by high cavity finesse and high average powers requires careful design of the cavity mirror coatings and precise control of the coating process.

In this letter, we address these challenges, focusing on the visible spectral range, using a recently developed widely tunable frequency comb source [28] and a novel cavity coating design. The design of our femtosecond enhancement cavity (fSEC) is shown in Fig. 1(a). The fSEC is a 4-mirror bow-tie configuration with 2 plane partial reflectors, M1 and M4, of nominally 0.3% transmission and 2 high reflectors (transmission < 0.1%), M2 and M3, with 50 cm radius of curvature. The mirrors are designed for operation from 450 to 700 nm. In order to minimize net
To characterize the fsEC performance, we first measure the cavity finesse, $F$, across the design wavelength range. To measure the finesse independent of the complications of the comb cavity coupling described below, we use the cavity attenuated phase shift (CAPS) method [14, 15]. In this method, the input laser to the cavity is amplitude modulated at an angular frequency $\Omega$ and the relative phase shift of the cavity transmitted light is measured. The phase shift, $\phi$, is due to the cavity acting as a low pass filter and is related to the cavity storage time, $\tau$, by:

$$\tan(\phi) = -\frac{\Omega}{\pi \tau}$$  \hspace{1cm} (1)

Which, in turn, is related to finesse via:

$$F = 2\pi \tau f_{\text{rep}}$$  \hspace{1cm} (2)

To measure finesse via the CAPS method while remaining frequency-locked to the cavity, we send two identical beams into the cavity in counterpropagating directions as shown in Fig. 1(a) and described in [14]. The forward beam, labeled signal, is used for locking while the second, labeled reference, is used for the measurement. For our CAPS implementation, the reference beam is sent through a Pockels cell between two polarizers. The Pockels cell is driven with sine wave with a peak value $\approx 3\%$ of the half-wave voltage. The first polarizer is detuned from the second, which optically biases the cell to get a cleaner sinusoidal output at the drive frequency. The reference beam at the cavity output is sent to a photodiode and lock-in amplifier measuring the phase shift relative to a leakage reference beam which bypasses the cavity.

The measured cavity finesse across the design range is shown in Fig. 1(c). The mean value is 927. The data shown is an average of 2 CAPS phase measurements taken at 70 and 90 kHz modulation frequency. At each frequency, the lock-in phase is recorded for 10 seconds and then the average and standard deviation is calculated which is used to find the error in each measurement. The error is higher on the long wavelength side which will be discussed below. As expected, the cavity finesse varies by more than a factor of 2 across the design range, with a general trend of increasing finesse at longer wavelengths. This is most likely due to absorption of the bluer side of the spectrum in the mirror coatings. The original design specification was for a finesse of $\sim 1000$ which is satisfied for most of the range. The cavity loss is dominated by the two PR mirrors and the cavity is nearly impedance matched [33]. In this case the absorption sensitivity enhancement factor remains approximately $F/\pi$ and is greater than 190 across the tuning range [34].
The CAPS measurements are relatively simple and independent of the details of the comb/cavity coupling. For measuring the performance of the setup for cavity-enhancing frequency combs with maximum bandwidth, more discussion of the frequency comb and stabilization schemes is required. For all these measurements, we use a two-point Pound-Drever-Hall (PDH) locking scheme [35], with a fast servo loop tightly locking one portion of the frequency comb to the cavity, and a second slower loop bringing another part of the comb onto resonance with a linearly independent actuator. This basic scheme has been used previously in many contexts [1, 14, 36, 37]. However, accomplishing comb/cavity coupling over such a large and unprecedented tuning range presents technical challenges, which we discuss in detail below.

The input combs are derived from a home-built synchronously pumped optical parametric oscillator (OPO) operating at 100 MHz repetition rate, previously described in [28]. This OPO is pumped by the second harmonic of a home-built high-power frequency comb laser consisting of a dispersive-wave shifted Er:fiber comb [25] with high-bandwidth transducers, amplified in a Yb-doped photonic crystal fiber amplifier [27]. To cover the entire tuning range, we use all three of the frequency doubled signal (2s, 450-515 nm), residual pump (535 nm), and doubled idler (2i, 555-700 nm) from the OPO. The phase transfer properties of the OPO, studied in detail in [28], necessitate different comb/cavity stabilization schemes for each of these combs. The schemes for 2i and 2s combs are shown in Fig. 2.

For coupling the 2i comb to the cavity, since phase modulation on the pump is transferred directly to the 2i comb with no bandwidth penalty imposed by the OPO cavity [28], we use an electro-optic modulator (EOM) in the mode-locked fiber laser to apply both 5 MHz PDH sidebands and feedback in the fast PDH loop, as shown in Fig. 2(a). This acts on the pump and 2i combs with a fixed point [38] near DC, mainly acting on the \( f_{\text{rep}} \) degree of freedom of the combs. The cavity’s free spectral range is kept within the EOM’s \( f_{\text{rep}} \) tuning range with an additional slow servo acting on the enhancement cavity length. For controlling the second comb degree of freedom in the slow PDH loop, we actuate on the OPO cavity length, which acts only on the comb’s \( f_0 \) degree of freedom. When coupling the pump light to the enhancement cavity, the fast loop is identical to the 2i case shown in Fig. 2(a) and the slow loop actuates on a temperature of the EOM in the mode-locked fiber laser to control \( f_0 \).

For coupling the 2s comb to the cavity, the high-speed actuators of the pump laser are of no use since phase modulation on the pump comb is not transferred to the 2s comb, as discussed in detail in [28]. Even though \( f_{\text{rep}} \) of the 2s comb must track that of the pump comb, these changes are offset by changes in the 2s comb’s \( f_0 \) such that the fixed point of the 2s comb with respect to pump phase modulation is near the 2s comb’s optical carrier frequency. Thus, comb/cavity coupling is accomplished with the pump comb free-running, and feedback signals are instead applied to the OPO cavity and fsEC, as shown in Fig. 2(b). The fast servo PDH loop drives a copper-bullet-style piezoelectric transducer (PZT) [39] to actuate on OPO cavity length and the \( f_0 \) of the 2s comb. PDH sidebands at 1.1 MHz are also generated by this PZT at a mechanical resonance. The slow loop controls the fsEC cavity length with a long travel PZT. When locking either the 2i or 2s combs, care must be taken when selecting the locking points in the spectrum since small changes in OPO cavity length can change the output spectrum dramatically which can result in complicated, multi-peaked intracavity spectra or significantly decreased power. Monitoring of the input and intracavity spectra simultaneously while locking helps avoid this problem.

The fundamental limit to the attainable intracavity pulse duration is given by the attainable intracavity spectral width, which is in turn related to the input spectrum and the intracavity dispersion. We have measured both the incident OPO input spectrum and intracavity spectra across the tuning range. Example intracavity spectra of the 2s, 2i, and pump combs are shown in Figs. 3(a)-(c), along with the corresponding input spectra. Figure 3(d) shows the input and intracavity spectral widths (2\( \sigma \)) measured at 13 wavelengths across the tuning range. The attainable intracavity bandwidth is less than the input OPO bandwidth due to residual mirror dispersion. Due to GDD, the intracavity spectra are more square-shaped than the input so standard Gaussian transform limit relations do not apply. Still, even the smallest bandwidths measured support sub-200 fs FWHM pulses across
the tuning range when Fourier transformed. In order to use the cavity-enhanced combs for our target application of ultrasensitive ultrafast optical spectroscopy [14, 15], low noise on the intracavity light is required. We have previously measured the amplitude noise of the input combs and found it to be sufficient [28]. However, since an optical cavity is a phase discriminator [33, 40], residual phase noise present on the input comb is converted to amplitude noise on the intracavity light. The data is shown in Fig. 4 for the 2s, 2i, and pump combs. The OPO idler inherits the phase noise from the pump and signal such that the 2i comb has the highest phase noise [28] of the three combs and thus the largest high-frequency RIN. Conversely, the 2s comb has the lowest optical phase noise and thus the lowest high-frequency RIN, despite far inferior servo bandwidth for the PZT locking scheme of Fig. 2(b).

In conclusion, we have for the first time demonstrated the cavity-enhancement of frequency combs with a widely tunable platform. The wide tuning range of > 7900 cm⁻¹ covers nearly the entire visible spectrum. Despite the technical challenges wavelength tuning imposes on the frequency comb generation, cavity mirrors, and comb/cavity coupling, we have demonstrated performance that is comparable to that used in previous experiments using cavity-enhanced combs [5, 14, 41]. For example, comparing to the previous one-wavelength demonstration of cavity-enhanced transient absorption spectroscopy [14], we report here the achievement that used in previous experiments using cavity-enhanced combs [5, 14, 41].

Acknowledgments. M.C. Silfies acknowledges support from the GAANN program of the U.S. Dept. of Education. G. Kowzan acknowledges support from the National Science Centre, Poland scholarship 2017/24/T/ST2/00242. The authors thank S.A. Diddams, H. Timmers, A. Kowligy, A. Lind, F.C. Cruz, N. Nader, and G. Ycas for assistance with Er comb development and providing manuscript feedback.

Disclosures. The authors declare no conflicts of interest.

REFERENCES

1. R. Jason Jones, I. Thomann, and J. Ye, Phys. Rev. A 69, 051803 (2004).
2. T. Gherman and D. Romanini, Opt. Express 10, 1033 (2002).
3. A. K. Mills, T. J. Hammond, M. H. C. Lam, and D. J. Jones, J. Phys. B: At. Mol. Opt. Phys. 45, 142001 (2012).
4. A. Cingöz, D. C. Yost, T. K. Allison, A. Ruehl, M. E. Fermann, I. Hartl, and J. Ye, Nature 482, 68 (2012).
5. C. Corder, P. Zhao, J. Bakalis, X. Li, M. D. Kershis, A. R. Muraca, M. G. White, and T. K. Allison, Struct. Dyn. 5, 054301 (2018).
6. A. K. Mills, S. Zhdanovich, M. X. Na, F. Boschin, E. Razzoli, M. Michiardi, A. Sheyerman, M. Schneider, T. J. Hammond, V. Süss, C. Felser, A. Damascelli, and D. J. Jones, Rev. Sci. Instruments 90, 083001 (2019).
7. T. Saule, S. Heinrich, J. Schöz, N. Lilienfein, M. Högner, O. de-Vries, M. Plötner, J. Weitenberg, D. Esser, J. Schulte, P. Russbueldt, J. Limpert, M. F. Kling, U. Kleineberg, and I. Pupeza, Nat. Commun. 10, 458 (2019).
8. C. Benko, L. Hua, T. K. Allison, F. m. c. Labaye, and J. Ye, Phys. Rev. Lett. 114, 153001 (2015).
9. R. Krischek, W. Wieczorek, A. Ozawa, N. Kiesel, P. Michelberger, T. Udem, and H. Weinfurter, Nat Photon 4, 170 (2010).
10. F. Adler, M. J. Thorpe, K. C. Cossel, and J. Ye, Annu. Rev. Anal. Chem. 3, 175 (2010).
11. G. Kowzan, D. Charczu, A. Cygan, R. S. Trawiński, D. Lisak, and P. Masłowski, Sci. Reports 9, 1 (2019).
12. P. B. Changala, M. L. Weichman, K. F. Lee, M. E. Fermann, and J. Ye, Science 363, 49 (2019).
13. C. A. Alrahman, A. Khodabakhsh, F. M. Schmidt, Z. Qu, and A. Foltynowicz, Opt. Express 22, 13889 (2014).
14. M. A. Reber, Y. Chen, and T. K. Allison, Optica 3, 311 (2016).
15. T. K. Allison, J. Phys. B: At. Mol. Opt. Phys. 50, 044004 (2017).
16. A. Schliesser, N. Pique, and T. W. Hansch, Nat Photon 6, 440 (2012).
17. H. Timmers, A. Kowligy, A. Lind, F. C. Cruz, N. Nader, M. Silfies, G. Ycas, T. K. Allison, P. G. Schunemann, S. B. Papp, and S. A. Diddams, Optica 5, 727 (2018).
18. M. Seidel, X. Xiao, S. A. Hussain, G. Arisonh, A. Hartung, K. T. Zawilski, P. G. Schunemann, F. Habel, M. Trubetskov, V. Pervak, O. Pronin, and F. Krausz, Sci. Adv. 4 (2018).
19. N. Leindecker, A. Marandi, R. L. Byer, K. L. Vodopyanov, J. Jiang, I. Hartl, M. Fermann, and P. G. Schunemann, Opt. Express 20, 7042 (2012).
20. K. F. Lee, J. Jiang, C. Mohr, J. Bethge, M. E. Fermann, N. Leindecker, K. L. Vodopyanov, P. G. Schunemann, and I. Hartl, Opt. Lett. 38, 1191 (2013).
21. A. Ruehl, A. Gambetta, I. Hartl, M. E. Fermann, K. S. E. Eikema, and M. Marangoni, Opt. Lett. 37, 2232 (2012).
22. G. Soñoñ, T. Martynikien, P. Mergo, L. Rutkowski, and A. Foltynowicz, Opt. Lett. 42, 1748 (2017).
23. T. Steinle, F. Mórz, A. Steinmann, and H. Giessen, Opt. Lett. 41, 4863 (2016).
24. T. Steinle, A. Steinmann, R. Hegenbarth, and H. Giessen, Opt. Express 22, 9567 (2014).
25. D. L. Maser, G. Ycas, W. I. Depetri, F. C. Cruz, and S. A. Diddams, Appl. Phys. B 123, 142 (2017).
26. A. Catanese, J. Rutledge, M. Silfies, X. Li, H. Timmers, A. S. Kowligy, A. Lind, S. A. Diddams, and T. K. Allison, arXiv:191203525 (2019).
27. X. Li, M. A. Reber, C. Corder, Y. Chen, P. Zhao, and T. K. Allison, Rev. Sci. Instruments 87, 093114 (2016).
28. Y. Chen, M. C. Silfies, G. Kowzan, J. M. Bautista, and T. K. Allison, Appl. Phys. B 125, 81 (2019).
29. F. Kaertner, L.-J. Chen, and G. Chang, U.S. patent 8,976,447 B2 (May 3, 2011).
30. A. Siegmans, Lasers (University Science Books, 1986).
31. J. M. Herbelin, J. A. McKay, M. A. Kwok, R. H. Ueunten, D. S. Urevig, D. J. Spencer, and D. J. Benard, Appl. Opt. 19, 144 (1980).
32. R. Engelin, G. von Helden, G. Berden, and G. Meijer, Chem. Phys. Lett. 262, 105 (1996).
33. W. Nagourney, Quantum Electronics for Atomic Physics and Telecommunication (OUP Oxford, 2014).
34. G. Gagliardi and H.-P. Loock, eds., Cavity-Enhanced Spectroscopy and Sensing, Springer Series in Optical Sciences (Springer-Verlag, Berlin Heidelberg, 2014).
35. R. W. P. Drever, J. L. Hall, F. V. Kowalski, J. Hough, G. M. Ford, A. J. Munley, and H. Ward, Appl. Phys. B: Lasers Opt. 31, 97 (1983).
36. A. Foltynowicz, P. Masłowski, A. J. Fleischer, B. J. Bjork, and J. Ye, Appl. Phys. B 110, 163 (2013).
37. C. Corder, P. Zhao, J. Bakalis, X. Li, M. D. Kershis, A. R. Muraca, M. G. White, and T. K. Allison, ProcSPIE 10519, 10519 (2018).
38. N. R. Newbury and W. C. Swann, J. Opt. Soc. Am. B 24, 1756 (2007).
39. T. C. Briles, D. C. Yost, A. Cingöz, J. Ye, and T. R. Schibli, Opt. Express 18, 9739 (2010).
40. M. Zhu and J. L. Hall, J. Opt. Soc. Am. B 10, 802 (1993).
41. N. Lilienfein, C. Hofer, M. Högner, T. Saule, M. Trubetskov, V. Pervak, E. Fill, C. Riek, A. Leitenstorfer, J. Limpert, F. Krausz, and I. Pupeza, Nat. Photonics 13, 214 (2019).
