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Spectral broadening of 2-mJ femtosecond pulses in a compact air-filled convex–concave multi-pass cell

Alan Omar,* Tim Vogel, Martin Hoffmann, and Clara J. Saraceno

Photonics and Ultrafast Laser Science, Ruhr-Universität Bochum, Universitätsstrasse 150, 44801 Bochum, Germany
Corresponding author: alan.omar@ruhr-uni-bochum.de

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Multi-pass cell (MPC) based temporal pulse compressors have emerged in recent years as a powerful and versatile solution to the intrinsic issue of long pulses from Yb-based high-power ultrafast lasers. The spectral broadening of high-energy (typically more than 100 µJ) pulses has only been realized in gas-filled MPCs due to the significantly lower nonlinear coefficient of gases compared with solid-state media. Whereas these systems reach impressive performance in terms of spectral broadening with very low spatiotemporal couplings, they are typically complex setups, i.e., large and costly pressure-controlled vacuum chambers to avoid strong focusing, ionization, and damage to the mirrors. Here, we present spectral broadening of 2-mJ pulses in a simple and compact (60-cm-long) multi-pass cell operated in ambient air. Instead of the traditional Herriott cell with concave-concave (CC/CC) mirrors, we use a convex–concave (CX/CC) design, where the beam stays large at all times, both minimizing damage and allowing operation in ambient air. We demonstrate spectral broadening of 2.1-mJ pulses at 100 kHz repetition rate (200 W of average power) from 2.1 nm (pulse duration of 670 fs) to a spectral bandwidth of 24.5 nm, supporting 133-fs pulses with 96% transmission efficiency. We show the compressibility of these pulses down to 134 fs and verify that the spectral homogeneity of the beam is similar to previously reported CC/CC designs. To the best of the authors' knowledge, this is the first report of a CX/CC MPC compressor operated at high pulse energies in air. Because of its simplicity, small footprint, and low cost, we believe this demonstration will have significant impact in the ultrafast laser community.

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High-average-power ultrafast lasers at 1 µm wavelength based on ytterbium-doped gain media have seen tremendous progress in the last decade [1–3], with average power levels largely exceeding the kilowatt level and increasingly widespread commercial availability. However, in comparison with this progress, their adoption in many applications and particularly in scientific research has been slow, due to the lack of efficient and simple paths to temporally reduce the typically long pulse durations of >300 fs to the realm of Ti:sapphire lasers, which commonly operate well below 100 fs, even with multi-millijoule pulse energies. For many years, gas-filled hollow-core capillaries [4] or filament compressors [5] were the mainstream techniques for nonlinear spectral broadening of millijoule-class pulses via self-phase modulation (SPM) in noble gases and subsequent pulse compression using dispersive delay lines. Using this technique, both a high average power and mJ energies have been successfully compressed down to few-cycle pulses. Some recent state-of-the-art demonstrations of the aforementioned techniques are a two-stage compression with gas-filled capillaries [6] and compression with a self-trapping filament in air [7]. In the first example, 240-fs pulses at 660 W of average power were compressed to 6.3 fs and 216 W with an optical transmission of 32% using Ne gas in the second stage at 7.5 bar. In the second example, 200-fs pulses were compressed at 4 mJ in a self-channeling region of 4.6 m to 40 fs. However, these techniques suffer from low efficiency, which is typically in the 50% range, and their practical implementation is cumbersome due to the complexity of the compression setup. Since their first demonstration in 2016 [8], Herriott-type multi-pass cells (MPC) have been recognized as a powerful technique to spectrally broaden and compress pulses with an extremely high throughput of >90% and a very wide range of input parameters and wavelength regions [9–11]. The main operation principle of spectral broadening in MPCs is to avoid beam degradation and catastrophic beam collapse by dividing the nonlinear spectral broadening into small enough steps with sufficient free-space propagation in between, which suppresses spatiotemporal coupling [12]. This combines the advantages of free-space propagation—which is critical for high average power lasers—while preserving excellent beam quality, even with large broadening factors. In pursuit of this goal, the concave-concave CC/CC Herriott-type MPC design is a practical design to introduce these multiple steps in a compact footprint with small misalignment sensitivity [13]. The nonlinear medium used to obtain spectral broadening can be a solid-state material (for example, fused silica) for low-energy systems (typically <100 µJ), or the gas itself inside the cell for higher-pulse-energy systems, with different scaling and design laws. So far, only the CC/CC Herriott-cell design has been used for pulse energies of >100 µJ, with a noble gas such as Ar, Kr, or Xe used as a nonlinear medium. In this way, remarkable results have been achieved; for instance, pulses from a 1-kW Yb:fiber laser system with a pulse duration of 200 fs were compressed to 31 fs at 1 mJ and an optical transmission of 96% in an argon-filled MPC in a 1.5-m-long chamber [14]. A similar technique was used by Kaumanns *et al*. to compress pulses with 18 mJ of pulse energy from 1.3 ps to 41 fs in a low-pressure chamber with a length of 3 m [15]. Balla *et al*. compressed 1.2-ps and 2-mJ pulses to a pulse duration of 32 fs with 1.6 mJ in a 2.5-m-long Kr-filled MPC with an optical transmission of 80% [16]. Recently, Pfaff *et al*. spectrally broadened 840-fs, 10-mJ pulses and showed compressibility to 38 fs with a high optical transmission of 97% and a low-pressure chamber length of 2.7 m [17]. While these results are certainly very impressive, they can only be achieved in a CC/CC MPC if critical parameters are carefully controlled. In 2022, Heyl *et al*. theoretically introduced a bow-tie MPC as a novel energy-scaling compression scheme and simulated the compression of 125 mJ from 1-ps pulses to 50 fs using a compact 2-m-long setup [18]. Another pulse energy scaling approach was introduced in Ref. [19]: spectral broadening via plasma-induced SPM in a conventional gas-filled MPC. According to simulations, this scheme can scale up to 10 J and compress 500-fs pulses down to 38 fs with a cavity length of 25 m [19]. In scaling the energy in the MPC, the naturally occurring focus in the CC/CC MPC has to be kept reasonably large. Otherwise, one can easily exceed the maximum nonlinear phase shift per pass that avoids spatiotemporal coupling, or the high peak intensity can give rise to gas ionization and subsequent beam degradation and pulse distortion. So far, this issue has been addressed by scaling the length of the gas chamber and the radii of curvature of the MPC mirrors, leading to impractical and expensive compression setups several meters in length [20].

Here, we present the use of a compact convex–concave (CX/CC) MPC in ambient air, i.e., without any vacuum or high-pressure chamber at 2.1 mJ pulse energy. This was possible with a carefully tailored MPC design using fast 3D pulse propagation simulations. As a first proof-of-principle result, we show nonlinear spectral broadening of 2.1-mJ, 670-fs pulses at 210 W of average power from 2.1 nm to 24.5 nm, corresponding to a Fourier-transform limit (FTL) of 133 fs. We show compressibility of the pulses down to 134 fs with comparable homogeneity to previously demonstrated designs at this high pulse energy. Furthermore, our air-filled cell is 60 cm long, and only requires standard highly-reflective mirrors for spectral broadening. We believe this is an important demonstration with respect to achieving more widely accessible Yb-doped high-power systems with short pulses. Our experiment was limited by the damage threshold of the available optics at the time of the experiment. We believe that pulse energy scaling to significantly higher energies beyond hundreds of mJ can be achieved by, on the one hand, operating at moderate vacuum levels to keep the peak power below the critical power, and on the other hand by decreasing the cavity length and increasing the radius of curvature of the CX/CV MPC to prevent damage to the MPC mirrors while additionally providing advantages in terms of compactness. The general guidelines for energy scaling in this modified MPC geometry require a thorough numerical exploration, which is currently the topic of a separate study using our fast 3D code and will be published elsewhere. We note that, at the time of the review of this paper, a demonstration of the CX/CC concept presented here at lower pulse energy (<200 μJ) was reported, which uses both fused silica and noble gas as the nonlinear medium and has 90% efficiency and excellent spatio-spectral homogeneity [21], adding additional proof of the versatility of the concept for many different laser parameters.

The driving laser system is a commercially available Yb-doped thin-disk regenerative amplifier (Trumpf Dira 500-10) with a maximum average power of 500 W which is operated at a repetition rate of 100 kHz, corresponding to a pulse energy of 5 mJ, and a constant pulse duration of 670 fs with a spectral width of 2.1 nm. The beam profile quality M^2 at the laser's output is 1.25×1.28 (in X and Y), with a slightly asymmetric profile. Using these parameters as input, we studied possible designs of CX/CC cells using numerical simulations. Driven by constraints in the available choice of CX mirrors in our laboratory at the time of the experiment (radius of curvature: −2.5 m), we studied an air-based MPC compressor using our home-made 3D pulse propagation tool. Our numerical model solves the spatiotemporal nonlinear Schrödinger equation in three dimensions (x, y, t) using the split-step Fourier method and considers linear effects (diffraction and dispersion) and nonlinear effects (optical Kerr effect, self-steepening, and instantaneous and delayed Raman scattering). The delayed Raman response is taken into account in air at room temperature (295 K) and, in this model, air is considered a mixture of 80% of N_2 and 20% of O_2 at atmospheric pressure [22]. In order to optimize the design and access a wide range of parameters, the code is written in C++ and Radeon Open Compute (ROCm) and is executed on a graphics processing unit (GPU) to reduce the computational time of one simulation. Moreover, the nonlinear refractive index of air in our simulation is 2.6×10^{-23} m^2/W, as measured by Schwarz *et al.* at a wavelength of 1054 nm and a pulse duration of 540 fs using a wavefront sensor technique [23].

For the design of our CX/CC MPCs, we used a 2-inch plano-CC mirror with a ROC of 2 m, and for the second mirror a 2-inch plano-CX mirror with a ROC of −2.5 m. The mirrors were separated by 61.5 cm, and we used 15 full roundtrips, corresponding to 30 reflections on either of the MPC mirrors. Both mirrors are high reflectors with close to zero group delay dispersion (GDD). The group velocity dispersion (GVD) of air at 1030 nm is 0.0162 fs^2/mm, which in our case results in a GDD per pass of 10 fs^2, and can therefore be neglected. In Fig. 1, the schematic of the experimental setup is shown. The beam of the amplifier is attenuated by a half-wave plate (HWP) and a thin-film polarizer (TFP). Two spherical mirrors are used to adjust the mode of the input beam to the necessary beam caustic as simulated using our 3D code, which allows for controlled propagation through the MPC without reaching the damage threshold of the MPC mirrors. The

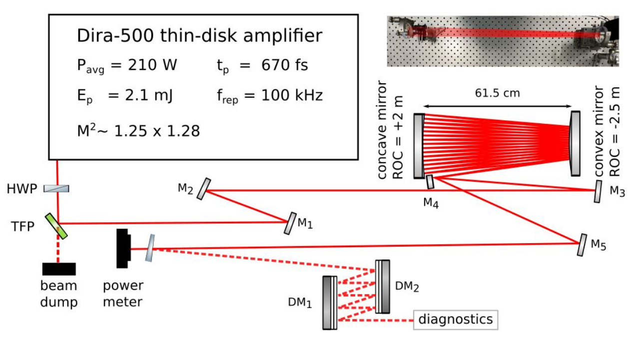

Fig. 1. Schematic of the setup. An Yb-doped thin-disk regenerative amplifier seeds the asymmetric air-filled CX/CC MPC. HWP: zero-order half-wave plate, TFP: thin-film polarizer, M1 and M2: curved HR mirrors are used for high-power mode matching, M4: in-coupling mirror, M5: recollimation mirror, PM: power meter, DM: dispersive mirrors.

beam then propagates through the MPC in a closed-path configuration where the beam is coupled in and out via a rectangular mirror. The estimated beam size ($1/e^2$) on the 7-mm rectangular in-coupling mirror is 1.65 mm, which limits the number of spots on the 2-inch CC mirror in our experiment, consequently preventing more nonlinear spectral broadening by increasing the number of passes. We are therefore, in this specific experiment, limited to a spectral broadening of a factor of approximately 10 (from 2.1 nm to 24.5 nm); however, future experiments with other adapted optical elements should allow shorter pulses to be reached. We note that, in spite of being operated in air, our simulations do not predict a significant influence of Raman scattering down to pulse durations of 50 fs. The total broadening (×10), and B-integral per pass in our setup (0.23 rad) were moderate compared with other values in the literature obtained using CC/CC designs; however, our simulations show that this same design can reach sub-50-fs pulses and a B-integral per pass of 0.6 rad without beam degradation, although we could not reach this point due to damage to the high-reflectivity, low-dispersion coating (GDD < 30 fs^2) of the CX mirror available at the time of the experiment, which was observed at a fluence of 0.3 J/cm^2. A formal comparison of the limits in terms of the B-integral per pass and a comparison between the CC/CC and CC/CX designs will be part of our upcoming detailed theoretical investigation. Even though we have high average power, we have not observed a significant temperature rise on the mirror coating or the optomechanical elements, as measured by a thermal camera. The MPC setup is placed in a housing to reduce the beam fluctuations due to air turbulence; however, the housing is not sealed in this case. The output beam is collimated with a spherical mirror, and the maximum output power is 201 W, which results in a power transmission efficiency of 96%. In our cell, the peak power is slightly below the critical peak power for self-focusing in air [23], as required in gas-filled MPCs [24]. For significantly higher peak powers, a slightly low-pressure, but still very compact, air-filled chamber can be easily implemented. The reflections on the MPC mirrors are the main cause of the residual power loss observed. At the time of the experiments, we did not have large dispersion optics to enlarge the beam sufficiently; therefore, to prevent damage to the dispersive mirrors, the power is attenuated with two silica wedges to 250 mW and compressed with 38 reflections on dispersive mirrors with a total GDD of −18 kfs^2. Our compressor was designed to compensate for GDD only, as higher-order terms only have minimal influence on our rather narrowband spectrum. The same compressibility is expected from the high-power beam given the availability of large enough dispersive optics. We characterized the compressed pulses using a home-built second-harmonic-generation (SHG) frequency-resolved optical gating (FROG) setup. Figures 2(a) and 2(b) show the results for the measured and retrieved traces of the compressed pulse. Both traces are in good agreement with each other, with a FROG error of 0.5% using a 512 × 512 grid. The blue-filled curve in Fig. 2(c) shows the measured spectrum obtained after the compression stage using an optical spectrum analyzer (OSA), which shows good agreement with the retrieved (dashed red) and simulated (dashed black) spectra; the green curve is the retrieved spectral phase of the compressed pulse. The central peak around 1030 nm in the measured spectrum relates to the amplifier spectrum and the characteristic pre- and post-pulses of the regenerative amplifier due to the leakage of the Pockels cell. These pulses do not contribute to the spectral broadening due to the extremely low energy in a

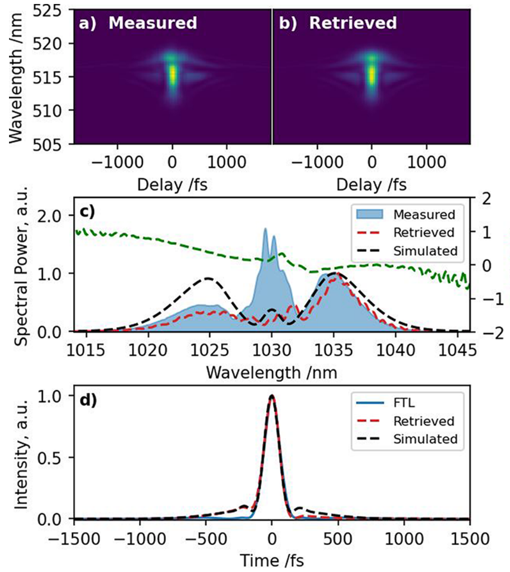

Fig. 2. Compression results measured with SHG-FROG. (a), (b) Measured and retrieved traces. (c) Measured, retrieved, and simulated spectra and spectral phase of the compressed pulse. (d) FTL, retrieved, and simulated temporal profiles of the pulse.

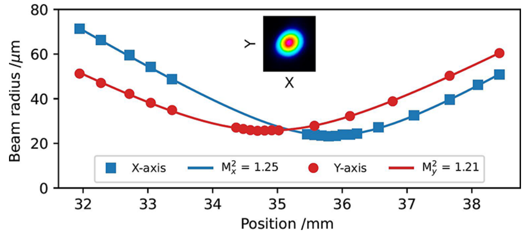

Fig. 3. Beam quality factor measurement (M^2) of the output beam along the transverse axes. The inset shows an image of the beam profile, as measured by a complementary metal-oxide semiconductor (CMOS) camera.

single side pulse, and the estimated deposited energy in these side pulses is 6%. In addition, amplified spontaneous emission (ASE) contributes approximately 1% of the energy deposited in the central lobe [14,17]. The calculated spectral bandwidth is 24.5 nm, corresponding to the Fourier-transform limit (FTL) of 133 fs. In Fig. 2(d), the retrieved temporal intensity profile (dashed red) is nearly transform limited (blue) and shows good agreement with the simulated pulse (dashed black). The compressed pulses have 18% of the pulse energy in the wings of the pulse. It exhibits a pulse duration at full width at half maximum (FWHM) of 134 fs with a compression factor of 5. Figure 3 depicts the good beam quality of the compressed pulse with a measured $M_x^2 \times M_y^2 = 1.25 \times 1.21$, which is comparable to the laser's output beam profile quality M^2 of 1.25 × 1.28. To investigate the spatio-spectral homogeneity of this configuration, we characterize the spectral homogeneity of the compressed beam by measuring the V-parameter [12]. Forty spectra are measured along the x axis and y axis of the collimated beam with a beam diameter ($1/e^2$) of $2w_x \times 2w_y = 2.6 \times 2.7$ mm by using a multimode fiber with a mode size of 200 μm (SMA connector) coupled into a spectrometer (Avantes Starline). The fiber is positioned on an XY-motorized stage for repeatable and accurate

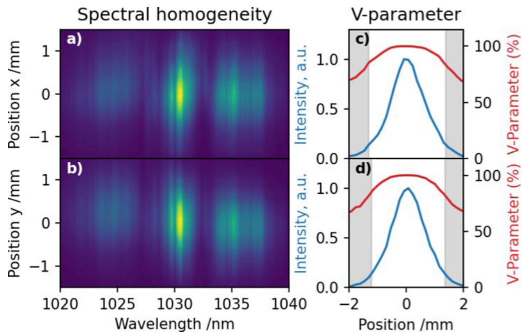

Fig. 4. Spatio-spectral homogeneity characterization of the compressed pulse for both axes. The solid blue curves on the right show the normalized intensity of the spectral profile. The red solid curves show the corresponding spatio-spectral homogeneity values (V-parameter), and the gray boxes show the beam area within a beam diameter ($1/e^2$).

measurements with a step size of 150 µm. Figures 4(a) and 4(b) depict the spectral homogeneity of both axes. In Figs. 4(c) and 4(d), the V-parameter (solid red) is over 82% (x axis) and 81% (y axis) within the $1/e^2$ beam area. The intensity-weighted average V-parameter is ∼90% for both axes within a 5-mm diameter of the measurement area. These results match the homogeneity measurements of other gas-filled Herriott-type MPCs, such as the V-parameter measured by Pfaff *et al.* [17].

In conclusion, we present, for the first time to the best of the authors' knowledge, the use of a CX/CC multi-pass cell in air for the spectral broadening of mJ pulses. We spectrally broadened the output pulses of a thin-disk regenerative amplifier from 670 fs, 2.1 mJ at 1030 nm and a high average power of 210 W (100 kHz repetition rate) to 134 fs and 203 W with an excellent optical efficiency of 96%, preserving the beam quality $M^2 < 1.3$ and obtaining a good spectral homogeneity. This novel scheme for asymmetric MPC represents a very compact and simple compressor to bring high-power and high-energy ultrafast lasers into the sub-100 fs regime and beyond. Compared with the more commonly utilized CC/CC MPC, it has the advantages of being much more compact and requiring ambient or—eventually—low air pressure, which can be achieved with very economic pre-pumps and sealed boxes. In the next step, we will extend this result to higher pulse energies of several tens of mJ (our laser system can provide up to 50 mJ at 10 kHz) and stronger spectral broadening by slightly reducing the air pressure, chirping the input pulses, and using optics with a higher damage threshold. According to our calculations, this could be achieved with very compact and simple cells such as the one presented here.

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

REFERENCES

1. F. Saltarelli, I. J. Graumann, L. Lang, D. Bauer, C. R. Phillips, and U. Keller, Opt. Express **27**, 31465 (2019).
2. J. Brons, V. Pervak, E. Fedulova, D. Bauer, D. Sutter, V. Kalashnikov, A. Apolonskiy, O. Pronin, and F. Krausz, Opt. Lett. **39**, 6442 (2014).
3. M. Müller, C. Aleshire, A. Klenke, E. Haddad, F. Légaré, A. Tünnermann, and J. Limpert, Opt. Lett. **45**, 3083 (2020).
4. M. Nisoli, S. D. Silvestri, O. Svelto, R. Szipöcs, K. Ferencz, C. Spielmann, S. Sartania, and F. Krausz, Opt. Lett. **22**, 522 (1997).
5. C. P. Hauri, W. Kornelis, F. W. Helbing, A. Heinrich, A. Couairon, A. Mysyrowicz, J. Biegert, and U. Keller, Appl. Phys. B **79**, 673 (2004).
6. S. Hädrich, M. Kienel, M. Müller, A. Klenke, J. Rothhardt, R. Klas, T. Gottschall, T. Eidam, A. Drozdy, P. Jójárt, Z. Várallyay, E. Cormier, K. Osvay, A. Tünnermann, and J. Limpert, Opt. Lett. **41**, 4332 (2016).
7. A. V. Mitrofanov, A. A. Voronin, D. A. Sidorov-Biryukov, G. Andriukaitis, T. Flöry, A. Pugžlys, A. B. Fedotov, J. M. Mikhailova, V. Y. Panchenko, A. Baltuška, and A. M. Zheltikov, Opt. Lett. **39**, 4659 (2014).
8. J. Schulte, T. Sartorius, J. Weitenberg, A. Vernaleken, and P. Russbueldt, Opt. Lett. **41**, 4511 (2016).
9. C.-L. Tsai, F. Meyer, A. Omar, Y. Wang, A.-Y. Liang, C.-H. Lu, M. Hoffmann, S.-D. Yang, and C. J. Saraceno, Opt. Lett. **44**, 4115 (2019).
10. P. Gierschke, C. Grebing, M. Abdelaal, M. Lenski, J. Buldt, Z. Wang, T. Heuermann, M. Mueller, M. Gebhardt, J. Rothhardt, and J. Limpert, Opt. Lett. **47**, 3511 (2022).
11. V. Hariton, A. B. Wahid, G. Figueira, K. Fritsch, and O. Pronin, Opt. Lett. **47**, 1246 (2022).
12. J. Weitenberg, A. Vernaleken, J. Schulte, A. Ozawa, T. Sartorius, V. Pervak, H.-D. Hoffmann, T. Udem, P. Russbüldt, and T. W. Hänsch, Opt. Express **25**, 20502 (2017).
13. D. Herriott, H. Kogelnik, and R. Kompfner, Appl. Opt. **3**, 523 (1964).
14. C. Grebing, M. Müller, J. Buldt, H. Stark, and J. Limpert, Opt. Lett. **45**, 6250 (2020).
15. M. Kaumanns, V. Pervak, D. Kormin, V. Leshchenko, A. Kessel, M. Ueffing, Y. Chen, and T. Nubbemeyer, Opt. Lett. **43**, 5877 (2018).
16. P. Balla, A. B. Wahid, and I. Sytcevich, *et al.*, Opt. Lett. **45**, 2572 (2020).
17. Y. Pfaff, C. Forster, G. Barbiero, M. Rampp, S. Klingebiel, J. Brons, C. Y. Teisset, H. Wang, R. Jung, J. Jaksic, A. H. Woldegeorgis, C. J. Saraceno, and T. Metzger, Opt. Express **30**, 10981 (2022).
18. C. M. Heyl, M. Seidel, E. Escoto, A. Schönberg, S. Carlström, G. Arisholm, T. Lang, and I. Hartl, JPhys Photonics **4**, 014002 (2022).
19. F. Tavella and K. Mecseki, Phys. Plasmas **29**, 083902 (2022).
20. M. Kaumanns, D. Kormin, T. Nubbemeyer, V. Pervak, and S. Karsch, Opt. Lett. **46**, 929 (2021).
21. K. Fritsch, V. Hariton, K. Schwarz, N. Kovalenko, G. Figueira, and O. Pronin, "Spectral broadening in convex-concave multipass cells," arXiv, arXiv:2211.10241 (2022).
22. J.-F. Ripoche, G. Grillon, B. Prade, M. Franco, E. Nibbering, R. Lange, and A. Mysyrowicz, Opt. Commun. **135**, 310 (1997).
23. J. Schwarz, P. Rambo, M. Kimmel, and B. Atherton, Opt. Express **20**, 8791 (2012).
24. M. Hanna, X. Délen, L. Lavenu, F. Guichard, Y. Zaouter, F. Druon, and P. Georges, J. Opt. Soc. Am. B **34**, 1340 (2017).