Dispersion control over the ultraviolet–visible–near-infrared spectral range with HfO_2/SiO_2-chirped dielectric multilayers

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We report the first realization, to the best of our knowledge, of a chirped multilayer dielectric mirror providing dispersion control over the spectral range of 300–900 nm and the first use of hafnium oxide in a chirped mirror. The technology opens the door to the reliable and reproducible generation of monocycle laser pulses in the blue–violet spectral range, will benefit the development of optical waveform and frequency-comb synthesizers over the ultraviolet–visible–near-infrared spectral range, and permits the development of ultrabroadband-chirped multilayers for high-power applications.

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Advancing ultrashort laser pulse generation to the limit set by the oscillation cycle of light has been pursued ever since the discovery of lasers. Pulses comprising an ever-decreasing number of wave cycles allow more efficient exploitation of nonlinear optical effects[1] with implications as striking as the generation of single subfemtosecond light pulses.[2] Moreover, the controlled superposition of light frequencies extending over more than one octave opens, along with carrier-envelope phase control,[3–5] the way to shaping the subcycle (i.e., subfemtosecond) evolution of light fields in laser pulses and thereby to a new way of quantum control based on the light-force-directed charge in atoms, molecules, or solids.[6] In this Letter, we present a chirped multilayer mirror offering high reflectivity and controlled group-delay dispersion (GDD) over some 1.5 octaves spanning from ultraviolet (UV) to near-infrared (NIR) frequencies. This technology may become instrumental for the development of future ultrawideband optical waveform synthesizers.

There have been several approaches to generating monocycle optical pulses: (i) phase-coherent superposition of pulses from different laser sources[7,8]; (ii) phase-coherent synthesis of Raman sidebands by exploiting vibrational or rotational transitions in molecules[9,10]; (iii) multicolor optical parametric generation[11]; and last but not least, (iv) by means of supercontinuum generation based on self- or cross-phase modulation.[12–14] What all these techniques have in common is that they rely on some optical device that can be used to adjust the phase (and amplitude) of individual groups of frequencies independently. This has, so far, been implemented by separating the spectral components of a broadband signal in space[15] and addressing the spectral channels in the Fourier plane by a spatial light modulator (SLM) based on liquid crystals[15] or acoustic waves.[16] The concept can, in principle, be extended to bandwidths exceeding one octave,[17] however, the UV–IR absorption and the low damage threshold of SLMs restricts the technology from being scaled to bandwidths of several octaves and ever higher pulse energies. In this Letter, we demonstrate that chirped multilayer mirrors may provide a promising alternative for specific applications, such as monocycle pulse generation or wideband frequency-comb generation.

Chirped mirrors (CM) have been continuously improved ever since their invention in 1994.[18] Progress has been made in terms of bandwidth, losses, GDD, and the ability to compensate higher-order spectral phase errors introduced by optical components.[19–27] As a result of the efforts of several groups, by the turn of the millennium, CM-based optical systems have been capable of controlling broadband radiation over spectral ranges approaching an octave in the visible–NIR domain.[12,13,28–30]

Recently, we demonstrated a chirped multilayer mirror supporting sub-3 fs pulses carried at a wavelength of 600 nm.[31] This technological progress paves the way toward the generation of monocycle light pulses, wideband optical waveform synthesis, and frequency-comb generation from compact, user-friendly laser systems. Many applications motivate the extension of these capabilities into the UV spectral range. The dramatically increasing dispersion of optical materials toward the UV absorption bands prevented chirped dielectric multilayer technology from being extended into the UV spectral range so far. By using HfO_2 (Refs. 32 and 33) in combination with SiO_2 as the high- and low-index material, respectively, here we demonstrate successful design and realization of what is believed to be the first ultrabroadband-chirped dielectric multilayer with controlled dispersion and high reflectivity extending to wavelengths as short as 300 nm.

The complementary pair of HfO_2/SiO_2 multilayers were designed with a commercially available software (OPTILAYER[34]) using a needle optimization algorithm.[35] The target of the mirror design was determined by the parameters of a pulse compressor under preparation: (i) high reflectivity in the spectral range of 300–900 nm and (ii) a frequency-dependent group delay compensating 1 mm of fused silica and 1 m of air in several bounces between for the pair. The feasibility of covering this spectral range with a coherent supercontinuum was recently demonstrated.[14]

The refractive index of HfO_2 ($n_H \sim 2.05$ at $\lambda \sim 500$ nm) is much smaller than that of high-index materials previously used in the fabrication of chirped multilayers, such as Nb_2O_5 ($n_H \sim 2.35$) and TiO_2 ($n_H \sim 2.45$). Achieving a high reflectivity over a large bandwidth with such a low value of n_H presents a formidable challenge. Our optimized design consists of 83 alternating layers of HfO_2 and SiO_2 and exhibits a spectrally averaged reflectivity of 93% in the 300–900 nm spectral range and a nominal GDD of -20 fs^2 at the center wavelength of $\sim$450 nm (see Fig. 1). For the reason discussed above, this effective reflectivity is lower than that of the 1.5-octave mirror demonstrated recently over the range of 400–1200 nm by the use of Nb_2O_5 as the high-index material,[31] but we are confident that it does not constitute the ultimate performance of this type of mirrors.

The HfO_2/SiO_2 multilayers were sputtered on fused-silica substrates of standard optical quality with a magnetron reactive Helios sputtering machine (Leybold Optics).[25,31] Details about the optimization of the HfO_2/SiO_2 deposition process with magnetron sputtering will be reported in a forthcoming publication.[33] The transmittivity and group delay versus wavelength of the manufactured mirrors were measured with a spectrophotometer (Lamda-950, PerkinElmer) and a homemade white-light interferometer. The latter device covers the spectral range of 400–1100 nm and is capable of determining the GDD with an accuracy of $\sim$10 fs^2 and a spectral resolution of 5 nm. The spectral transmittivity measurements were supplemented reflectivity measurement at selected wavelengths to accurately determine the frequency-dependent mirror reflectivity. From these measurements, we obtained parasitic (scattering and absorption) losses to be not higher than 0.1% at 800 nm.

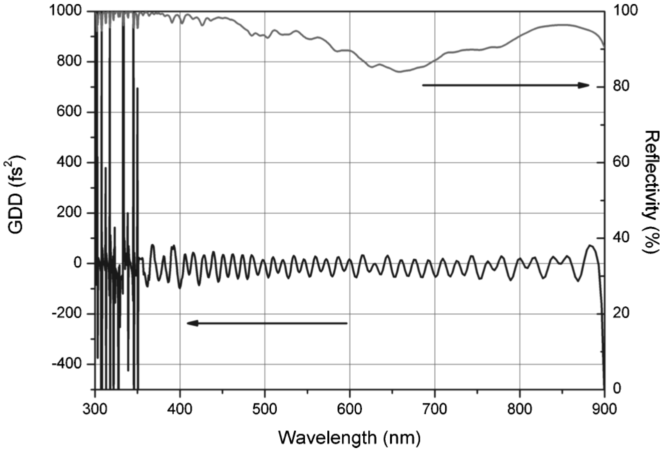

Fig. 1. Calculated averaged reflectivity (gray curve) and averaged GDD (black curve) for the optimized complementary mirror pair described in the text.

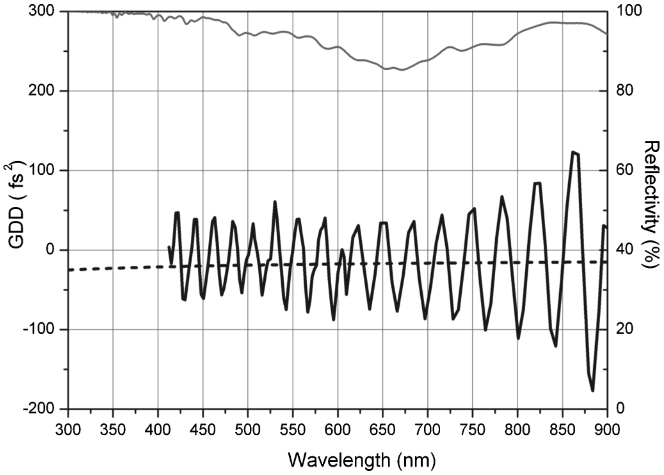

Fig. 2. Measured reflectivity (gray curve) and GDD (black solid curve) averaged for a complementary pair of chirped mirrors. The dashed curve depicts the design target for the mirror GDD.

Comparison of the reflectivity and dispersion data of the manufactured (Fig. 2) and designed (Fig. 1) mirrors reveal close agreement. The accurate reproduction of the calculated reflectivity over the entire design spectral range of 300–900 nm and of the mean (cycle-averaged) GDD over the range of 400–900 nm leaves no room for dramatic deviations of the GDD from the calculated behavior in the 300–400 nm spectral range, which cannot be accessed by our white-light interferometer at present.

To analyze the potential these mirrors offer for pulse compression and optical waveform synthesis, we have considered a hypothetical broadband dispersive delay line with five bounces off each of the two complementary-chirped mirrors (total GDD at 450 nm approximately -200 fs^2) compensating the dispersion of 2 mm fused silica and 0.4 m of air. The measured GDD curve over the 400–900 nm range was supplemented with the design curve over the range of 300–400 nm for the calculations. We have sent a hypothetical Gaussian pulse carried at a wavelength of 450 nm (and corresponding spectrum shown in the inset of Fig. 3) with a duration of 1.8 fs (FWHM) through this optical delay line and calculated the temporal intensity profile of the pulse exiting the system in the same way as in Ref. 31. The results are summarized in Fig. 3. The uncompensated spectral oscillations in the mirror's GDD curve leads

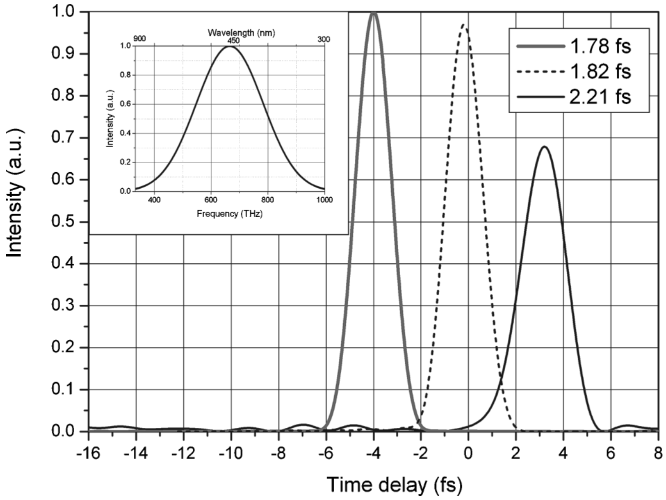

Fig. 3. Influence of a dispersive delay line made up of the new chirped mirrors on a 1.78 fs, 450 nm laser pulse. The gray and black curves depict the intensity profile of the incoming and exiting pulse, respectively (with an artificial delay). Dashed line: the pulse profile after 2 bounces.

to a broadening of the pulse to approximately 2.2 fs, accompanied by a decrease of the pulse energy by a factor of ~2. The fine structure that can be seen on the dispersion curve of the design at ~300–350 nm in Fig. 1 apparently does not severely affect the reflected pulse properties.[36]

In conclusion, we have reported the first extension of ultrabroadband dispersion control with chirped dielectric multilayers well into the UV spectral range. The demonstrated complementary pair of HfO_2/SiO_2-chirped multilayers offers the potential for monocycle pulse generation in the blue–violet spectral range and lends itself for applications in future ultrabroadband optical waveform and frequency-comb synthesizers. Thanks to the high damage threshold of HfO_2,[37] this work also opens the way for the development of a chirped multilayer for applications in high-power laser systems.

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References

1. T. Brabec and F. Krausz, Rev. Mod. Phys. **72**, 545 (2000).
2. M. Hentschel, R. Kienberger, Ch. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, Nature **414**, 509 (2001).
3. D. J. Jones, S. A. Diddams, J. K. Ranka, A. Stentz, R. S. Windeler, J. L. Hall, and S. T. Cundiff, Science **288**, 635 (2000).
4. A. Apolonski, A. Poppe, G. Tempea, C. Spielmann, T. Udem, R. Holzwarth, T. Hänsch, and F. Krausz, Phys. Rev. Lett. **85**, 740 (2000).
5. A. Baltuska, T. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, C. Gohle, R. Holzwarth, V. S. Yakovlev, A. Scrinzi, T. W. Hänsch, and F. Krausz, Nature **421**, 611 (2003).
6. M. F. Kling, Ch. Siedschlag, A. J. Verhoef, J. I. Khan, M. Schultze, Th. Uphues, Y. Ni, M. Uiberacker, M. Drescher, F. Krausz, and M. J. J. Vrakking, Science **312**, 246 (2006).
7. T. W. Hänsch, Opt. Express **80**, 71 (1990).
8. O. Albert and G. Mourou, Appl. Phys. B **69**, 207 (1999).
9. A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **85**, 562 (2000).
10. A. E. Kaplan, Phys. Rev. Lett. **73**, 1243 (1994).
11. Y. Kobayashi, H. Takada, M. Kakehata, and K. Torizuka, Appl. Phys. Lett. **83**, 839 (2003).
12. M. Nisoli, S. De Silvestri, O. Svelto, R. Szipöcs, K. Ferencz, Ch. Spielmann, S. Sartania, and F. Krausz, Opt. Lett. **22**, 522 (1997).
13. K. Yamane, Z. Zhang, K. Oka, R. Morita, M. Yamashita, and A. Suguro, Opt. Lett. **28**, 2258 (2003).
14. N. Aközbeck, S. A. Trushin, A. Baltuska, W. Fuss, E. Goulielmakis, K. Kosma, F. Krausz, S. Panja, M. Uiberacker, W. E. Schmid, A. Becker, M. Scalore, and M. Bloemer, New J. Phys. **8**, 177 (2006).
15. A. M. Weiner, Rev. Sci. Instrum. **71**, 1929 (2000).
16. F. Verluise, V. Laude, Z. Cheng, Ch. Spielmann, and P. Tournois, Opt. Lett. **25**, 575 (2000).
17. T. Binhammer, E. Rittweger, U. Morgner, R. Ell, and F. X. Kärtner, Opt. Lett. **31**, 1552 (2006).
18. R. Szipöcs, K. Ferencz, C. Spielmann, and F. Krausz, Opt. Lett. **19**, 201 (1994).
19. F. X. Kärtner, N. Matuschek, T. Schibli, U. Keller, H. A. Haus, C. Heine, R. Morf, V. Scheuer, M. Tilsch, and T. Tschudi, Opt. Lett. **22**, 831 (1997).
20. N. Matuschek, L. Gallmann, D. H. Sutter, G. Steinmeyer, and U. Keller, Appl. Phys. B **71**, 509 (2000).
21. G. Tempea, V. Yakovlev, B. Bacovic, F. Krausz, and K. Ferencz, J. Opt. Soc. Am. B **18**, 1747 (2001).
22. V. Yakovlev and G. Tempea, Appl. Opt. **41**, 6514 (2002).
23. G. Steinmeyer, Opt. Express **11**, 2385 (2003).
24. G. Tempea, V. Yakovlev, F. Krausz, in *Springer Series in Optical Sciences*, N. Kaiser and H. Pulker, eds. (Springer, 2003).
25. V. Pervak, S. Naumov, G. Tempea, V. Yakovlev, F. Krausz, and A. Apolonski, Proc. SPIE **5963**, 490 (2005).
26. J. Kim, J. R. Birge, V. Sharma, J. G. Fujimoto, F. X. Kärtner, V. Scheuer, and G. Angelow, Opt. Lett. **30**, 1569 (2005).
27. G. Steinmeyer, Appl. Opt. **45**, 1484 (2006).
28. P. Dombi, V. S. Yakovlev, K. O'Keeffe, T. Fuji, M. Lezius, and G. Tempea, Opt. Express **13**, 10888 (2005).
29. G. Steinmeyer and G. Stibenz, Appl. Phys. B **82**, 175 (2006).
30. M. Yamashita, K. Yamane, and R. Morita, IEEE J. Sel. Top. Quantum Electron. **12**, 213 (2006).
31. V. Pervak, A. V. Tikhonravov, M. K. Trubetskov, S. Naumov, F. Krausz, and A. Apolonski, Appl. Phys. B (published online: DOI 10.1007/s00340-006-2467-8).
32. H. A. Macleod, *Thin Film Optical Filters*, 3rd ed. (Institute of Physics, 2001).
33. V. Pervak, F. Krausz, and A. Apolonski, Thin Solid Films (to be published).
34. OptiLayer software: http://www.optilayer.com.
35. A. V. Tikhonravov, M. K. Trubetskov, and G. W. DeBell, Appl. Opt. **35**, 5493 (1996).
36. A. V. Tikhonravov, Ph.D. dissertation (Physics Faculty, Moscow State University, 1986).
37. R. Chow, S. Falabella, G. E. Loomis, F. Rainer, C. J. Stolz, and M. R. Kozlowski, Appl. Opt. **32**, 5567 (1993).