Ultrabroadband chirped mirrors for femtosecond lasers

E. J. Mayer, J. Möbius, A. Euteneuer, and W. W. Rühle

Department of Physics, Philipps University, Renthof 5, D-35032 Marburg, Germany

R. Szipőcs

R&D Lézer-Optika Bt., P.O. Box 622, H-1539 Budapest, Hungary

Received November 25, 1996

We report on the performance of widely tunable femtosecond and continuous-wave Ti:sapphire lasers that use a newly developed ultrabroadband mirror set. The mirrors exhibit high reflectivity ($R > 99\%$) and smooth variation of group delay versus frequency over a wavelength range from 660 to 1060 nm. Mode-locked operation with pulse durations of 85 fs was achieved from 693 to 978 nm with only one set of ultrabroadband mirrors. © 1997 Optical Society of America

Tunable laser sources are widely used for cw and time-resolved spectroscopic measurements.[1] In particular, cw, picosecond, and femtosecond (fs) Ti:sapphire (Ti:S) lasers[2] have become versatile tools in spectroscopy because of the large bandwidth of the Ti:S crystal of 3200 cm^{-1} (FWHM),[3] which allows a broad tunability range as well as the generation of ultrashort pulses by self-mode locking of the laser.[4] The limitations on the minimum pulse durations of prism-pair-controlled lasers due to third- and fourth-order dispersion have been considerably reduced by the development of chirped dispersive dielectric mirrors[5,6] (CM's). In contrast with prism-pair-controlled systems, these mirrors compensate not only the second-order dispersion, often referred to as group-delay dispersion (GDD), but also the third-order dispersion. Up to now, CM's have been developed solely as a means of designing ultrafast laser systems with pulses as short as possible[7,8] or to simplify cavity design and improve the efficiency of fs laser systems.[9,10] In this Letter we demonstrate that our newly developed ultrabroadband CM's (UBCM's) offer the possibility of tuning ultrabroadband fs laser systems without changing mirrors over the whole of the laser's operation range. These UBCM's enable us not only to generate pulses shorter than 100 fs from a mode-locked Ti:S system but also simultaneously to achieve high tunability of the laser over a range >300 nm.

The problem of designing ultrabroadband dielectric mirrors for fs laser systems is twofold. First, the mirrors have to have continuous high reflectivity over a broad spectral range without any drop in reflectivity regardless of wavelengths. Second, the mirrors have to exhibit a smooth, possibly negative variation of the group-delay versus frequency function over the whole tuning range, allowing fs mode-locked operation of the laser. The present commercially available broadband dielectric mirrors do not meet these requirements. The high-reflectivity bandwidth of our preferred UBCM's can be exceeded only with properly designed metallic mirrors with similarly smooth variation of group delay versus frequency; however, the reflectivity of metallic mirrors is considerably smaller than that of UBCM's and therefore metallic mirrors cannot be used as intracavity broadband mirrors in fs laser oscillators. Here we show that the two requirements are fulfilled by one solution: CM structures[5,6] with an increasing layer period toward the substrate of the coating. Additionally, the mirrors have to be transparent at the pump wavelength(s) to replace standard [quarter-wave ($\lambda/4$) stack] dichroic mirror coatings[11] in these fs laser cavities. Note that it is impossible to fulfill the latter requirement with metallic mirrors.

In the case of dielectric mirrors, a combination of materials with the highest refractive-index ratios (n_H/n_L) is usually preferred since the higher the ratio, the higher the theoretical reflectance and bandwidth of standard $\lambda/4$ stacks. Among its competitors, the $\mathrm{TiO_2/SiO_2}$ pair has the highest ratio over the near-IR spectral range.[11] To produce a high-density coating with low scattering and absorption losses, ion-based technologies could be advantageous.[12] However, the total number of layers is strictly limited by the relatively high stress in such coatings, which does not allow the deposition of CM's formed by a relatively high number of thick layers in the near IR.[13] In the case of Ti:S lasers, for instance, the useful bandwidth of low-dispersion quarter-wave mirrors is limited to ~180 nm around 800 nm.[11] Previously proposed solutions to extend the high-reflectivity range of dielectric mirrors, such as deposition of low- (high-) pass stacks as a single coating[14] and deposition of multilayer stacks with variation of thickness in arithmetic or geometric progression[15] do not meet the above-listed requirements.[11,16–18] Briefly, all the previously used broadband dielectric mirrors exhibited rapid change of the reflected phase at specific wavelengths in the high-reflectivity zone of the broadband mirrors, causing resonant losses and extremely strong high-order dispersions around these wavelengths, thus preventing their use in broadly tunable fs oscillators.[11] Recently, it was demonstrated that these undesirable resonant features are effectively eliminated if their design is optimized by special computer algorithms.[6,19] From the theoretical point of view, we showed that it is possible to synthesize extremely broadband dielectric high reflectors that exhibit smooth, monotonic variation of group delay versus frequency throughout the complete high-

0146-9592/97/080528-03$10.00/0

reflectivity range of the mirrors and are transparent for the pump wavelengths.[6] The resulting gradient-index structure exhibits an increasing layer period toward the substrate, i.e., chirped multilayer structure, high reflectivity, and negative GDD over most of the fluorescence band of Ti:S.

These recent advances in deposition[9,11] and coating-design techniques[5,6,19] paved the way for the development of UBCM's for broadly tunable cw and ultrafast Ti:S lasers. Figure 1 shows the calculated transmittance of one of our state-of-the-art UBCM's. A high-reflectivity ($R > 99\%$) range from 660 to 1060 nm that covers most of the fluorescence band of the Ti:S was obtained by computer optimization.[5,19] The mirrors are designed for high transmission ($T > 90\%$) at the pump wavelengths of 488 and 514 nm to test the UBCM's in a fs Ti:S laser system (Coherent MIRA 900) pumped by a multiline 8.0-W Ar^+ laser (Coherent Innova 400). Further improvement in the pump efficiency could be achieved either by use of a single-line green pump source, e.g., a cw, intracavity frequency-doubled $Nd{:}YVO_4$ laser or by improvement of the present mirror design.

This specific design is built with alternating layers of SiO_2 and TiO_2 as low- and high-index materials, respectively, with optical thicknesses varying around 200 nm, one-fourth of our selected wavelength regime. Optical-thickness coefficients of the design are given in the caption for Fig. 1. Further technical details on the coating-deposition technology are available in Ref. 11. The theoretical smooth variation of group delay versus frequency of the UBCM's is plotted in Fig. 1. We verified the dispersive properties of the CM coatings after the deposition process by using the white-light interferometric technique.[20] The mirrors are designed to have an average negative GDD of $-50\ \text{fs}^2$ and a positive third-order dispersion of $+75\ \text{fs}^3$ around 800 nm to ensure nearly ideal dispersive conditions for mode-locked operation. The considerable extension of the high-reflectivity range of the CM's with respect to previous designs[5,7,8,10] was achieved at the expense of a slightly higher fluctuation in the negative GDD that, however, does not affect the formation of pulses longer than 50 fs: The theoretical GDD curve includes values between -20 and $-80\ \text{fs}^2$ over most of the tuning range, with a slight oscillation around the average value.

To demonstrate the performance of the UBCM's, we replaced all mirrors, including the dichroic pump mirror, with UBCM's, except for the output coupler (OC) in our Ti:S laser. The spectra of the Ti:S output pulses are characterized by an optical spectrum analyzer and the pulse width by intensity and fringe-resolved autocorrelation (FRAC) measurements with a 0.5-mm-long KTP crystal.

The Ti:S laser is continuously tunable from 681 to 1013 nm in cw operation with only one change of the OC. The linewidth of the cw laser is <0.15 nm when a two-plate birefringent filter wavelength-selecting element is used. One standard mirror, the OC, with a reduced reflection band has to be used in the laser solely to prevent laser activities at higher orders of the birefringent filter. The wavelength dependence of the cw output power is similar to that in mode-locked operation (Fig. 2), with a maximum of 1.34 W at 770 nm. The wavelength range of cw operation agrees quite well with the calculated high reflectivity range of the UBCM's (Fig. 1) except at very long wavelengths. We recall that, in practice, the features of dielectric mirrors do not fit their ideal (theoretical) reflectance–transmittance characteristics perfectly because of losses in the layers. In particular, the lower gain of the Ti:S crystal at longer wavelengths makes the system more sensitive to reflection losses on the mirrors. Thus we attribute the long-wavelength cutoff of the lasing activity to a slightly smaller reflectivity of the UBCM's with respect to standard $\lambda/4$ stack mirrors. Note that the UBCM's are expected to have the highest losses on the long-wavelength side of their reflectivity range, since in this wavelength range their group delay has the highest values in proportion to reflection losses.[11]

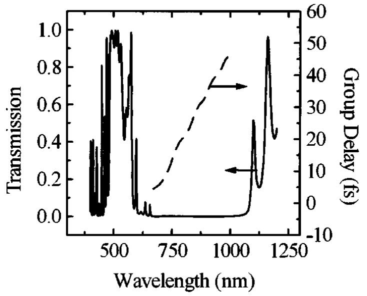

Fig. 1. Transmittance (solid curve) and group delay (dashed curve) of an UBCM versus wavelength. Optical thickness coefficients of the design are[19] $S|1.31L\ 1.70H\ 1.43L\ 0.66H\ 1.55L\ 1.45H\ 1.04L\ 1.20H\ 1.14L\ 1.32H\ 1.47L\ 0.99H\ 0.97L\ 1.17H\ 1.46L\ 1.15H\ 1.18L\ 1.11H\ 1.09L\ 1.08H\ 1.11L\ 1.33H\ 1.19L\ 0.91H\ 1.11L\ 0.96H\ 1.05L\ 0.83H\ 0.93L\ 1.11H\ 1.01L\ 0.98H\ 0.85L\ 0.90H\ 0.79L\ 0.99H\ 0.80L\ 0.93H\ 0.96L\ 0.60H\ 0.69L\ 1.09H\ 0.97L\ 0.41H\ 0.59L\ 1.35H\ 0.90L\ 0.10H|A$, where S is substrate, $n_S = 1.51$; A is air, $n_A = 1.0$; and H and L are $\lambda/4$ layers of TiO_2 and SiO_2, respectively, at $\lambda = 790$ nm, with $n_H = 2.315$, $n_L = 1.45$.

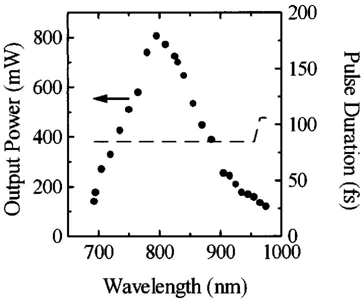

Fig. 2. Output power (dots) and pulse duration (dashed line) of the fs Ti:S laser with UBCM's. For intracavity dispersion control, a standard Brewster-angled prism pair made of SF10 glass was used in a Coherent MIRA 900 laser with a prism separation of 60 cm.

Figure 2 shows the measured output power over the tuning range (693 to 975 nm) in mode-locked operation. The plotted output power was measured while the pulse duration was kept constant at ~85 fs. In practice, we were not able to observe any decrease of

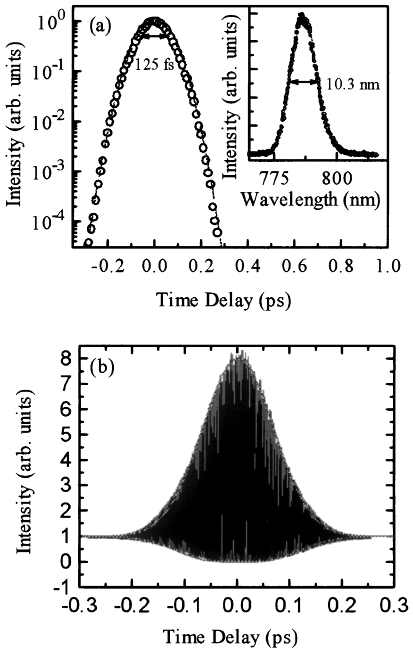

Fig. 3. (a) Autocorrelation curve (open circles) of the pulses at 805 nm (logarithmic scale). Dashed curve, Gaussian fit to the pulse shape. Inset: spectrum of the pulse (linear scale). (b) FRAC trace at $\lambda = 789$ nm ($\Delta\lambda = 10.3$ nm).

the laser output power when using the UBCM's instead of $\lambda/4$-stack standard-type mirrors. The intracavity negative GDD required for mode-locked operation is provided by a standard Brewster-angled prism pair and the UBCM's. This hybrid solution is necessary since the negative GDD of practicable UBCM's is too low to compensate for the positive-material GDD of the 20-mm-long Ti:S crystal used in our laser setup. The hybrid dispersion-control system combines the advantages of the CM approach, which allows higher-order dispersion compensation for relatively long crystals, and continuous variation of the GDD by the prisms. This means that we are able to keep the pulses transform limited at a pulse duration of 85 fs over the whole tunability range as well as to achieve pulse durations as short as 53 fs if we also use an external-prism-pair pulse compressor.

In Fig. 3(a), we plot the intensity autocorrelation trace and the spectrum (inset) at $\lambda = 786$ nm without using any extracavity GDD control. The dashed curves are fits to a Gaussian pulse shape. From the pulse duration of 89 fs and the bandwidth of 10.3 nm we calculate a time–bandwidth product of 0.44, which is in good agreement with the value for Gaussian transform-limited pulses. Except for the pulses obtained at the limits of the tuning range (i.e., for $\lambda < 730$ nm and $\lambda > 930$ nm) we measure a nearly constant value of the time–bandwidth product of 0.44 (note, however, that by removing the two-plate birefringent filter from the cavity, we obtained clear, nearly transform-limited sech^2 pulses). The transform-limited nature of the pulses has been proved by a FRAC trace [Fig. 3(b)]. The fringes of the FRAC traces are clearly resolved even in the wings, and no substructure is observed in the envelopes.

We have demonstrated the outstanding performance of our new ultrabroadband chirped mirrors in a broadly tunable cw and fs Ti:S laser. We achieved continuous tunability of the Ti:S laser with pulse durations of 85 fs for a wavelength range of 280 nm by changing only the OC. The tunability of the present setup is limited because of the use of standard OC's. After determining the optimum operation conditions, we plan to design a single broadband OC with dispersive properties similar to those of the UBCM's to develop fully tunable, user-friendly cw and ultrafast lasers.

Finally, the broadband mirrors presented here can also be adapted to other commercially available cw and ultrafast laser systems containing active materials with spectrally broad fluorescence[9] or nonlinear materials for parametric-wave generation.[10] Similar UBCM's exhibiting high reflectivity and smooth variation of the group delay versus frequency might be well suited for compressing optical pulses well below 5 fs.[8]

The authors thank K. Ferencz for manufacturing the mirrors. This research was supported by the Science Foundation of Hungary under grants CW-015285 and T-02056, and by R&D Lézer-Optika Bt., Budapest, Hungary.

References

1. W. Demtröder, *Laser Spectroscopy*, 2nd ed. (Springer-Verlag, Berlin, 1996).
2. J. D. Kafka, M. L. Watts, and J. W. J. Pieterse, IEEE J. Quantum Electron. **28,** 2151 (1992).
3. P. F. Moulton, J. Opt. Soc. Am. B **3,** 125 (1986).
4. D. E. Spence, P. N. Kean, and W. Sibbett, Opt. Lett. **16,** 42 (1991).
5. R. Szipőcs, K. Ferencz, Ch. Spielmann, and F. Krausz, Opt. Lett. **19,** 201 (1994).
6. R. Szipőcs and A. Kőházi-Kis, Proc. SPIE **2253,** 140 (1994).
7. L. Xu, Ch. Spielmann, F. Krausz, and R. Szipőcs, Opt. Lett. **21,** 1259 (1996).
8. A. Baltuska, Z. Wei, M. S. Pshenichnikov, and D. A. Wiersma, Opt. Lett. **22,** 102 (1997).
9. I. T. Sorokina, E. Sorokin, E. Wintner, A. Cassanho, H. P. Jenssen, and R. Szipőcs, Opt. Lett. **21,** 1165 (1996).
10. J. Hebling, E. J. Mayer, J. Kuhl, and R. Szipőcs, Opt. Lett. **20,** 919 (1995).
11. For a review, see, e.g., K. Ferencz and R. Szipőcs, Opt. Eng. **32,** 2525 (1993).
12. P. J. Martin, J. Mater. Sci. **21,** 1 (1986).
13. C. S. Masser, Newport Corporation, P.O. Box 19607, Irvine, Calif. 92713 (personal communication, 1995).
14. A. F. Turner and P. W. Baumeister, Appl. Opt. **5,** 69 (1966).
15. O. S. Heavens and H. M. Liddell, Appl. Opt. **5,** 373 (1966).
16. J. Ebert, H. Pannhorst, H. Küster, and H. Welling, Appl. Opt. **18,** 818 (1979).
17. P. Laporta and V. Magni, Appl. Opt. **24,** 2014 (1985).
18. A. M. Weiner, J. G. Fujimoto, and E. P. Ippen, Opt. Lett. **10,** 71 (1985).
19. R. Szipőcs and A. Kőházi-Kis, "Theory and design of chirped mirrors," submitted to Appl. Phys. B.
20. A. P. Kovács, K. Osvay, Zs. Bor, and R. Szipőcs, Opt. Lett. **20,** 788 (1995).