



Complementary Si/SiO₂ dispersive mirrors for 2-4 μm spectral range

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Abstract: Complementary pair of dispersive multilayers operating in the 2-4 μm spectral range were designed and produced for the first time. The mirrors comprise layers of Si and SiO₂ thin-film materials. The pair exhibits unparalleled reflectance exceeding 99.7% and provides a group delay dispersion of (-200) fs². The mirrors can be used in Cr:ZnS/Cr:ZnSe femtosecond lasers and amplifiers.

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1. Introduction

Dispersive mirrors (DM) are being actively used for dispersion compensation within laser systems and manipulation of ultrashort pulses in a number of laser applications [1–5]. The DM technology has become already well-established in the systems based on Ti:Sa lasers (~800 nm) and near-infrared Yb:YAG lasers (~1030 nm). The next step was done by pushing the DM optical elements into Thulium- and Holmium-based lasers systems operating around 2 μm [6,7]. The novel technology based on 2.4 μm chromium doped zinc sulfide (Cr:ZnS) or chromium doped zinc selenide (Cr:ZnSe) will allow one to extend the laser output to 3.2 μm [8,9]. For further development of this technology, dispersive mirrors in the spectral range 2-4 μm are strongly demanded. Until now, the best achievement in this direction is the authors' recently published work [10], where two DMs operating in the range 2-3.2 μm were reported. The DMs provided high reflectance exceeding 99.6% and group delay dispersion (GDD) of (-100 fs²) and (-200 fs²). Due to well-optimized deposition process, the mirrors exhibited reduced O-H absorption around the wavelength of 2.7 μm . In the present work, the next step in the development of infrared dispersive optics was done: a complementary pair of DMs operating in the entire 2-4 μm spectral range was designed and produced. The pair provides reflectance higher than 99.7% and group delay dispersion (GDD) of (-200 fs²) over the 2-4 μm range. The mirrors comprise layers of the same materials as in [10], namely Si and SiO₂. Exploitation of the thin-film combinations typical for the visible-near-infrared spectral range (for example, Nb₂O₅/SiO₂, Ta₂O₅/SiO₂, TiO₂/SiO₂) cannot provide large negative GDD values required for the development of Cr:ZnS/Cr:ZnSe lasers since oscillations in GDD reach too high unacceptable values. The main difference from Ref. [10] is that the complimentary pair approach was developed and applied for Si/SiO₂ technology for the first time. Due to this approach, unprecedented large GDD values can be compensated in the Cr:ZnS/Cr:ZnSe lasers.

In Section 2, design process of the complementary DMs is described. In Section 3, production process and characterization results are shown, and benefits of the developed complimentary pair are demonstrated. Conclusions are presented in Section 4.

2. Design and simulation

It is known that due to interference between the waves reflected from the top layer and the wavelengths reflected from the subsequent deeper layers, obtaining DM design with absolutely flat GDD is not possible [11]. In well-optimized DMs, GDD oscillates around its average value. The GDD oscillations can broaden the pulse and lead to appearing so-called satellite pulses. The considered design problem is challenging because of a large bandwidth, high required reflectance values and negative GDD values of (-200 fs^2). In order to resolve this challenge, not one DM but a complementary DM pair [12] was designed. The complementary DM pair includes two DMs with GDD curves exhibiting oscillations of the same amplitude but shifted by a half period (see red and blue curves in Fig. 1(a)). As GDD of the pair is the sum of GDD of two DMs, the lowest possible oscillations can be achieved.

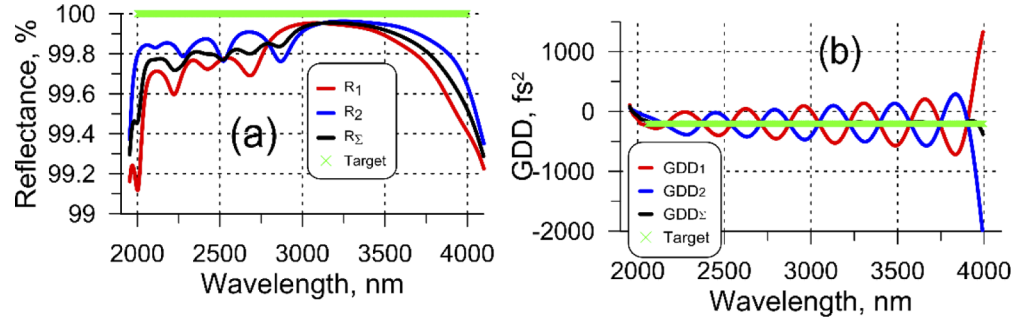


Fig. 1. (a) Reflectance R_1 , R_2 of the designed DMs, DM1851-S and DM1851-L, respectively. (b) Group delay dispersion, GDD_1 , GDD_2 , of the designed DMs, DM1851-S and DM1851-L, respectively. Calculated reflectance R_Σ (a) and GDD_Σ (b) of the DM1851 complimentary pair.

Nominal refractive indices of SiO_2 and Si were described by the well-known Cauchy formula:

$$n(\lambda) = A_0 + A_1 \left(\frac{\lambda_0}{\lambda} \right)^2 + A_2 \left(\frac{\lambda_0}{\lambda} \right)^4 \quad (1)$$

where A_0 , A_1 , A_2 are dimensionless parameters, $\lambda_0 = 1000 \text{ nm}$, λ is specified in nanometers. For SiO_2 $A_0 = 2.065721$, $A_1 = 0.016830$, $A_2 = 0.001686$, and for Si, $A_0 = 3.213463$, $A_1 = 0.150903$, $A_2 = -3.305936 \cdot 10^{-3}$ [10].

The DM designs were calculated with the help of the needle optimization method and a special multi-coating algorithm implemented into the OptiLayer software [12–14]. The design process was based on the minimization of the combined merit function comparing actual spectral performance of the complementary DM pair with target spectral characteristics. The combined merit function was defined as:

$$\begin{aligned} MF^2 &= \alpha_1 MF_1^2 + \alpha_2 MF_2^2 + MF_\Sigma^2, \\ MF_1^2 &= \sum_{j=1}^{1000} \left(\frac{R^{(p)}(\mathbf{X}^{(1)}; \lambda_j) - 100}{\Delta_{R,j}} \right)^2 + \sum_{j=1}^{1000} \left(\frac{GDD^{(p)}(\mathbf{X}^{(1)}; \lambda_j) + 200}{\Delta_{GDD,j}} \right)^2, \\ MF_2^2 &= \sum_{j=1}^{1000} \left(\frac{R^{(p)}(\mathbf{X}^{(2)}; \lambda_j) - 100}{\Delta_{R,j}} \right)^2 + \sum_{j=1}^{1000} \left(\frac{GDD^{(p)}(\mathbf{X}^{(2)}; \lambda_j) + 200}{\Delta_{GDD,j}} \right)^2, \\ MF_\Sigma^2 &= \sum_{j=1}^{1000} \left(\frac{R_\Sigma^{(p)}(\lambda_j) - 100}{\Delta_{R,j}} \right)^2 + \sum_{j=1}^{1000} \left(\frac{GDD_\Sigma^{(p)}(\lambda_j) + 200}{\Delta_{GDD,j}} \right)^2, \end{aligned} \quad (2)$$

where $\{\lambda_j\}$ are evenly distributed wavelength points in the spectral range of interest from 2000 nm to 4000 nm, $X^{(1)} = \{d_1^{(1)}, \dots, d_m^{(1)}\}$ and $X^{(2)} = \{d_1^{(2)}, \dots, d_n^{(2)}\}$ are the vectors of layer thicknesses of DMs of the complementary pair; $R^{(p)}(X^{(1)}; \lambda_j) = R_1$ and $R^{(p)}(X^{(2)}; \lambda_j) = R_2$ are actual reflectance of the first and the second mirrors, respectively; m and n are the layer counts in the mirrors, the angle of incidence is 5° , (p) denotes p-polarization; $\{\Delta_{R,j}\}$, $\{\Delta_{GDD,j}\}$ are design tolerances; α_1 , and α_2 are target weights corresponding to the first and second mirrors, respectively. Reflectance R_Σ and group delay dispersion GDD_Σ of the complementary pair are calculated as follows:

$$R_\Sigma = \sqrt{R^{(p)}(X^{(1)}) \cdot R^{(p)}(X^{(2)})}, \quad GDD_\Sigma = (GDD^{(p)}(X^{(1)}) + GDD^{(p)}(X^{(2)}))/2 \quad (3)$$

The design algorithms calculates analytically all derivatives, including first and second order derivatives of the merit function, which in their turn recall the analytical derivatives of the phase shift with respect to the angular frequency. These calculations provide the highest accuracy of the design algorithm.

As a result, two DMs denoted as DM1851-S and DM1851-L were synthesized. Spectral reflectance and GDD of both DMs as well as reflectance of the pair is presented in Fig. 1. The DM1851-S and DM1851-L designs comprise 28 layers. The design structures are presented in Fig. 2. The reflectance, GD and GDD of the designs are almost not sensitive to the incidence angles from 0° to 15° .

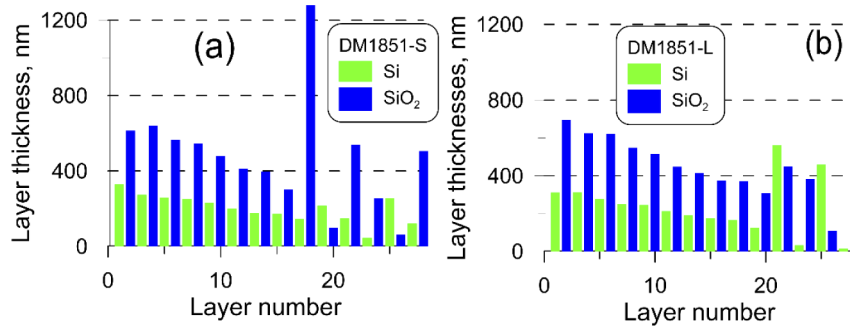


Fig. 2. Design structure of DM1851-S (a) and DM1851-L (b).

3. Production and characterization

The designed DMs were produced using a Helios deposition plant (Bühler Leybold Optics) based on magnetron sputtering, with the layer thicknesses controlled using well-calibrated time monitoring [12]. The plant is equipped with two proprietary dual-magnetrons and a plasma source for ion-assisted reactive middle frequency dual-magnetron sputtering. The magnetrons were optimized for high sputtering rates and high optical layer performance. The system was pumped by turbo-molecular pumps to 1×10^{-6} mbar before deposition. Argon and oxygen (for SiO_2 layers only) were used for both magnetrons. In the magnetron cathodes, Si target was used, the electric power of the cathode was 4500 W. The gas pressure was 1×10^{-3} mbar during the sputtering process. In the case of SiO_2 layers, oxygen was fed near the targets to oxidize the sputtering films. The distance from the targets to the substrates was 100 mm. The deposition rates were around 0.5 nm/s for both materials. Two samples, DM1851-S and DM1851-L, corresponding to eponymous designs were produced in two subsequent deposition runs. The deposition accuracy was of outmost importance since even small errors in layers thicknesses and refractive indices can cause shifting of the GDD spectral curves and degrade the expected perfect spectral GDD performance of the designed complimentary pair.

After the deposition, reflectance data of DM1851-S and DM1851-L samples was measured by the universal reflectance accessory (Perkin Elmer) in the range from 1400 nm to 3100 nm with the wavelength step of 2 nm (see red curves in Fig. 3), the measurement accuracy was 0.2%. The blue curve in Fig. 3 presents experimental reflectance recorded by Fourier transform infrared spectrometer Vertex 70 (Bruker Optics GmbH) in the wavelength range from 2000 nm to 4100 nm. In Fig. 3, an excellent correspondence between experimental data and theoretical reflectance data of both DMs is observed. The degradation of reflectance indicated around 2700 nm is pronounced in measurement data from both devices. This degradation is explained by inevitable presence of water vapor in the coatings and in the air.

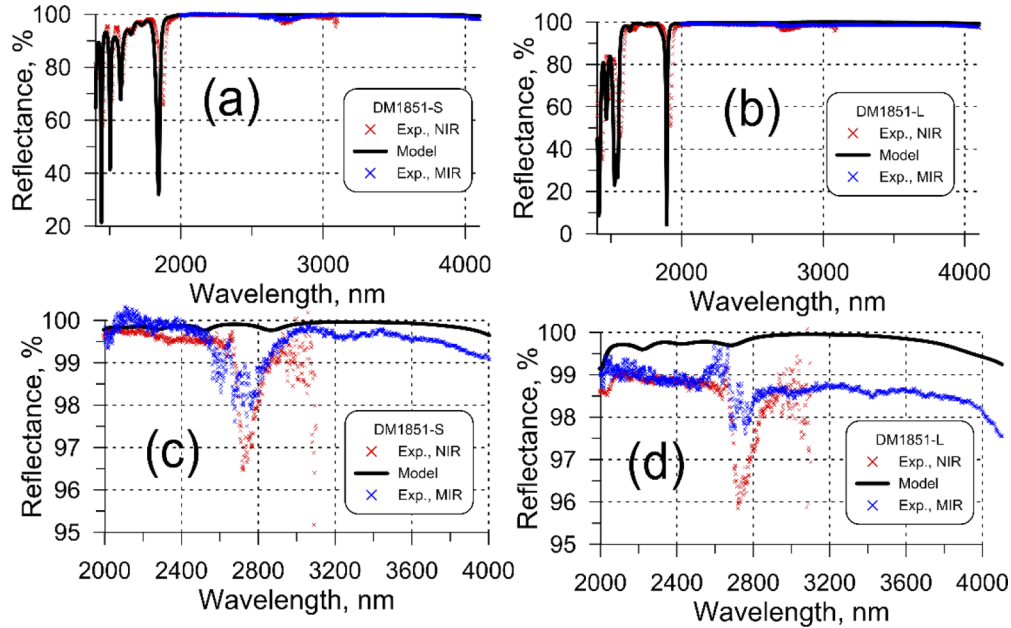


Fig. 3. Comparison of the nominal and experimental reflectance of the samples DM1851-S (a,c) and DM1851-L (b,d). Experimental data recorded in two different spectral ranges is shown by red (near-infrared range) and blue (mid-infrared range) crosses. Panel on the bottom show the data in the operating range from 2 to 4 μm .

The group delay (GD) and GDD experimental values of DM1851-S and DM1851-L were calculated from the data recorded by a mid-infrared white-light interferometer (WLI) developed in-house [15]. Theoretical and experimental GD/GDD values are compared in Fig. 4 and Fig. 5, respectively. The measurements show a good agreement with the theoretical curves for the GD as well as the GDD curves. Especially for the GDD all the specific maxima and minima could be measured for both mirrors. For DM1851-S there is a slight shift towards longer wavelengths while for DM1851-L the extrema are shifted slightly towards the shorter wavelengths. Below 3 μm the signal gets noisier due to the decreasing performance of the used detector in this range. O-H absorption around 2700 nm does not affect the GD/GDD measurements.

For designed DMs, the simulations of pulse propagation were performed [14]. In Fig. 6, the simulated envelopes of the input Fourier-limited pulse and pulse reflected from the designed DM1851 DMs are plotted. In the course of the simulations, it was assumed that the full width of half maximum of the input pulse is 55 THz and the central wavelength is 3000 nm. Total reflectance R_N and total GDD_N after N bounces in a laser setup will be:

$$R_N = R(DM_1) \cdot \dots \cdot R(DM_N), GDD_N = GDD_1 + \dots + GDD_N \quad (4)$$

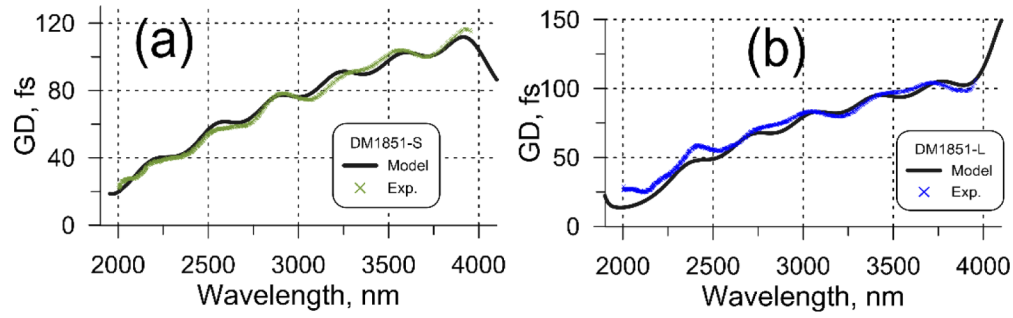


Fig. 4. Comparison of the nominal and experimental GD values of the samples DM1851-S (a) and DM1851-L (b).

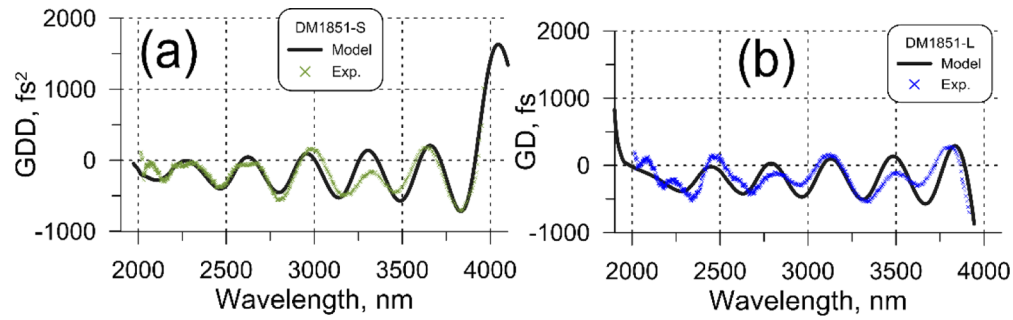


Fig. 5. Comparison of the nominal and experimental GDD values of the samples DM1851-S (a) and DM1851-L (b).

It is seen that after two bounces, the pulse does not change at all. With 16 bounces ($N = 16$, blue curve in Fig. 6), the complementary pair provides total reflectance of 98% and GDD of (-3200 fs^2). On the contrary, the single DM1851-S mirror exhibits large losses of about 20% with 16 bounces (see pink curve in Fig. 6). Also, appearance of satellite pulses is clearly observed.

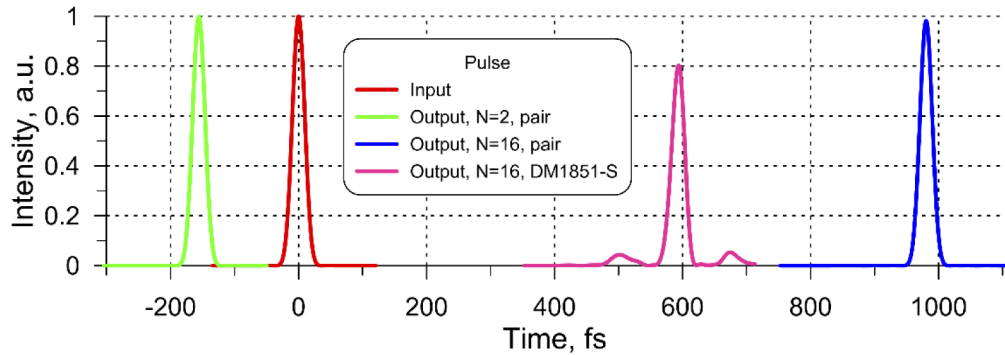


Fig. 6. Input and output pulse simulations calculated after different bounce count.

4. Conclusions

A one-octave complimentary pair of dispersive mirrors providing the group delay dispersion of (-200 fs^2) in the spectral range from 2-4 μm has been successfully designed and produced for

the first time. The mirrors comprise layers of Si and SiO₂ providing the large high-/low-index contrast and enabling achieving high reflectance and specified group delay dispersion over the entire broadband spectral range of interest. Unparalleled reflectance exceeding 99.7% and flat group delay dispersion in the 2-4 μm spectral range makes its possible exploiting the produced mirrors inside the Cr:ZnS/Cr:ZnSe femtosecond lasers and amplifiers and demonstrate high potential for further development of the mid-infrared multilayer optics.

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Disclosures

The authors declare no conflicts of interest.

References

1. V. Pervak, O. Razskazovskaya, I. B. Angelov, K. L. Vodopyanov, and M. Trubetskov, "Dispersive mirror technology for ultrafast lasers in the range 220–4500 nm," *Adv. Opt. Technol.* **3**(1), 1–9 (2014).
2. E. Fedulova, K. Fritsch, J. Brons, O. Pronin, T. Amotchkina, M. Trubetskov, F. Krausz, and V. Pervak, "Highly-dispersive mirrors reach new levels of dispersion," *Opt. Express* **23**(11), 13788 (2015).
3. P. Dombi, P. Rác, M. Lenner, V. Pervak, and F. Krausz, "Dispersion management in femtosecond laser oscillators with highly dispersive mirrors," *Opt. Express* **17**(22), 20598–20604 (2009).
4. V. Pervak, O. Pronin, O. Razskazovskaya, J. Brons, I. B. Angelov, M. K. Trubetskov, A. V. Tikhonravov, and F. Krausz, "High-dispersive mirrors for high power applications," *Opt. Express* **20**(4), 4503–4508 (2012).
5. V. Pervak, C. Teisset, A. Sugita, S. Naumov, F. Krausz, and A. Apolonski, "High-dispersive mirrors for femtosecond lasers," *Opt. Express* **16**(14), 10220–10233 (2008).
6. K. Yang, H. Bromberger, H. Ruf, H. Schäfer, J. Neuhaus, T. Dekorsy, C. V.-B. Grimm, M. Helm, K. Biermann, and H. Künzel, "Passively mode-locked Tm,Ho:YAG laser at 2 μ m based on saturable absorption of intersubband transitions in quantum wells," *Opt. Express* **18**(7), 6537 (2010).
7. T. Amotchkina, M. Trubetskov, F. Habel, V. Pervak, J. Zhang, K. Mak, O. Pronin, F. Krausz, and V. Pervak, "Synthesis, fabrication and characterization of a highly-dispersive mirrors for the 2 μ m spectral range," *Opt. Express* **25**(9), 10234 (2017).
8. S. Mirov, I. Moskalev, S. Vasilyev, V. Smolski, V. Fedorov, D. Martyshkin, J. Peppers, M. Mirov, A. Dergachev, and V. Gapontsev, "Frontiers of mid-IR lasers based on transition metal doped chalcogenides," *IEEE J. Sel. Top. Quantum Electron.* **24**(5), 1–29 (2018).
9. I. T. Sorokina and E. Sorokin, "Femtosecond Cr²⁺ based lasers," *IEEE J. Sel. Top. Quantum Electron.* **21**(1), 273–291 (2015).
10. V. Pervak, T. Amotchkina, Q. Wang, O. Pronin, K. F. Mak, and M. Trubetskov, "2/3 octave Si/SiO₂ infrared dispersive mirrors open new horizons in ultrafast multilayer optics," *Opt. Express* **27**(1), 55 (2019).
11. G. Steinmeyer, "Femtosecond dispersion compensation with multilayer coatings: toward the optical octave," *Appl. Opt.* **45**(7), 1484 (2006).
12. V. Pervak, A. V. Tikhonravov, M. K. Trubetskov, S. Naumov, F. Krausz, and A. Apolonski, "1.5-octave chirped mirror for pulse compression down to sub-3 fs," *Appl. Phys. B: Lasers Opt.* **87**(1), 5–12 (2007).
13. A. V. Tikhonravov, M. K. Trubetskov, and G. W. DeBell, "Optical coating design approaches based on the needle optimization technique," *Appl. Opt.* **46**(5), 704–710 (2007).
14. A. V. Tikhonravov and M. K. Trubetskov, "OptiLayer software," <http://www.optilayer.com>.
15. F. Habel, M. Trubetskov, and V. Pervak, "Group delay dispersion measurements in the mid-infrared spectral range of 2–20 μ m," *Opt. Express* **24**(15), 16705–16710 (2016).