

Empirical study of the group delay dispersion achievable with multilayer mirrors

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Abstract: With the help of the most advanced algorithms we obtained many dozens of multilayer dispersive mirror designs to empirically find limits for the maximum achievable negative value of the group delay dispersion (GDD). This value depends on the total thickness of coatings and layer material combination. Nb₂O₅/SiO₂ and Ta₂O₅/SiO₂ combinations are studied in detail, for combinations HfO₂/SiO₂ and TiO₂/SiO₂ we obtained estimations for two bandwidths. We also show that reasonable values of third-order dispersion have no significant impact on the obtained results. Current state-of-the-art technology allows to produce designs with total physical thicknesses slightly higher than 10 μm and to achieve maximum negative GDD values corresponding to this total design thickness. Designs with total physical thickness of 15 μm and 20 μm are not realized yet due to high sensitivity to deposition errors.

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OCIS Codes: (310.4165) Multilayer design; (320.5520) Pulse compression; (310.6805) Theory and design.

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1. Introduction

During the last 20 years dispersive mirrors (DM) technology [1–14] has become very popular in femto- and atto-seconds physics [15,16]. Nowadays the majority of femtosecond lasers include DM optics which provides an accurate GD/GDD control of different wavelength components. For example, precise GD/GDD control over an unprecedented bandwidth of almost two octaves allows one to generate pulses with the duration less than 2 fs in the visible range [16]. Therefore, the design of DM with required spectral performance (reflectance and group delay dispersion (GDD) characteristics) is crucial for many modern applications in the field of ultrafast physics.

Projecting of complicated ultrafast laser systems requires knowledge of possible DM properties that should be utilized during GD/GDD control in such systems. Currently there is high and rapidly rising demand on the DMs possessing various spectral properties and quite often designers of ultrafast laser systems are not aware of existing limitations. It makes the designing of ultrafast systems more complicated; quite often expensive resources and time are wasted because of the lack of this knowledge. For example, development of a DM compressor involves an important decision of a number of pulse bounces in DM system which can be estimated basing on a given input pulse bandwidth and phase modulation. Using the results of the presented work one can estimate maximum achievable levels of GDD per one pulse bounce. The required number of pulse bounces is easily estimated by dividing of input pulse GDD by this value.

DM optics with large bandwidth and large negative GDD is necessary for compensation of GDD accumulated by a pulse passed through dispersive media, optical amplifiers, etc. The larger bandwidth is required, the smaller the mean value of GDD can be achieved [6,7,11]. This fact seems to be caused by fundamental causality principle and is closely connected with phase properties of multilayers [17]. Further theoretical study of the relations between maximum achievable negative GDD values and reflectance might be interesting.

In this work we concentrate on a numerical study of possible DM designs. Our goal is to draw a limit of achievable GDD values as a function of total physical thickness of a DM and working bandwidth for different layer material pairs. GDD is the main parameter responsible for the temporal change of a pulse envelope. Third-order dispersion (TOD) and higher order dispersions can be easily taken into account in the design procedure by adjusting the shape of the target $GDD(\lambda)$ wavelength dependence.

Complementary pairs of DMs [14,18] and double-angle DMs [19] are out of the scope of the current work, while using these approached one can significantly decrease unavoidable GDD oscillations. The broadest 1.5 optical octave DM compression was achieved with complimentary pairs approach [18]. Nevertheless we consider single DM only, since in many cases it is used as a basic building block of more complicated systems.

2. Formulation of the design problem and solution approach

We used OptiLayer software [20] to design all coatings. The software utilizes needle optimization and gradual evolution techniques [21–23] that are the most powerful design approaches allowing to avoid problems of convergence to a local minimum. The results of the last Coating Design Contests [24–26] serve as an independent confirmation of this statement.

Target requirement specifications were as follows. At the normal incidence all designs should have reflectance higher than 99%. This level of reflectance provided more than 90% of energy in the pulse after 10 bounces in DM compressor. Additional requirement was that the pulse duration should remain close to Fourier transform limit for the corresponding bandwidth (allowed FWHM increase of the pulse <5%). In all cases the central wavelength was 800 nm (the central wavelength of Ti:Sapphire laser).

We concentrate on Nb_2O_5/SiO_2 and Ta_2O_5/SiO_2 combinations, since they are the most widespread for the production of DMs with the magnetron-sputtering technique. Usually Nb_2O_5/SiO_2 is used for broadband mirrors due to a larger contrast in refractive indexes. Ta_2O_5/SiO_2 is used for low loss coatings with relatively small bandwidths (200–300 nm). For Nb_2O_5/SiO_2 and Ta_2O_5/SiO_2 layer material combinations and for different values of the bandwidth and total physical/optical thickness we tried to obtain DM designs fulfilling above requirements and providing maximum possible negative GDD values in the specified spectral band. Combinations HfO_2/SiO_2 and TiO_2/SiO_2 were considered in more limited sense, just in order to provide the reader with general tendencies. The total number of designs obtained in this study exceeds 130.

Table 1. Cauchy formula coefficients for the substrate and layer materials

Material	n_∞	A	B	n at 800nm
Suprasil	1.443268	0.004060	6.94818E-6	1.4496
SiO_2	1.465294	0.0	4.71080E-4	1.4664
Nb_2O_5	2.218485	0.021827	3.99968E-3	2.2624
Ta_2O_5	2.065721	0.016830	1.686E-3	2.0961
TiO_2	2.317083	0.021242	6.60512E-3	2.3664
HfO_2	2.024462	0.015650	0.0	2.0489

In order to make the results more practical we used refractive indices of Nb_2O_5 , Ta_2O_5 , HfO_2 , and SiO_2 layer materials related to the magnetron-sputtering deposition process. These refractive indices were obtained as a result of a careful characterization of layer optical properties (depositions performed with Leybold Optics HELIOS coater). The combination HfO_2/SiO_2 has less contrast of refractive indices comparing to other combinations. Due to this

fact to achieve target specifications one needs significantly thicker designs. It makes sense to use this combination only when high intensities of light are to be expected, since this combination possesses better laser-induced damage threshold [27,28] or in cases when the working bandwidth spreads below 300 nm. We also included TiO₂ layer material, which is used quite often as a high index material in relatively simple chirped mirrors production. Unfortunately optical properties of TiO₂ layers deposited with the magnetron-sputtering process are not stable [29,30], making production of DMs with challenging characteristics much more complicated. Basing on these considerations we selected properties of TiO₂ layer material corresponding to ion-beam sputtering deposition process [31] providing dense and homogeneous TiO₂ films. In our study the refractive index spectral dependence is specified with the Cauchy formula $n(\lambda) = n_{\infty} + A/\lambda^2 + B/\lambda^4$, and corresponding coefficients are represented in Table 1 for all layer materials and Suprasil substrate (λ should be expressed in μm).

3. Nb₂O₅/SiO₂ layer materials pair

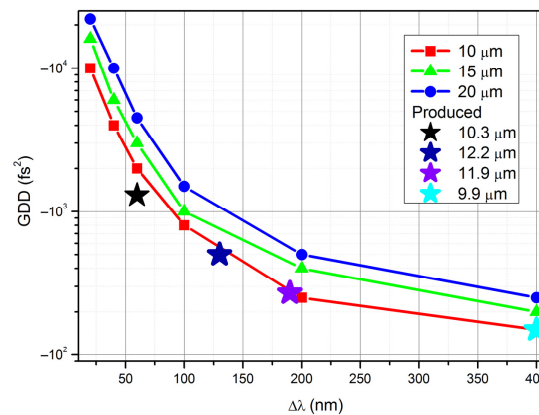


Fig. 1. Summarized main parameters of obtained designs with Nb₂O₅/SiO₂ layer materials pair. Red, green and blue curves correspond to designs with the maximum physical thickness of 10 μm , 15 μm and 20 μm , respectively. Asterisks correspond to produced designs [5,11,32] with physical thicknesses shown in the legend.

For Nb₂O₅/SiO₂ layer materials pair we designed DMs for the bandwidths of 20 nm, 40 nm, 60 nm, 100 nm, 200 nm, and 400 nm. As a termination condition for the gradual evolution procedure [22] we chose the total physical thickness on levels of 10 μm , 15 μm and 20 μm . Maximum achievable negative GDD values of designs versus the bandwidth are presented in Fig. 1. This figure demonstrates that maximum achievable negative GDD levels rapidly decrease with growing the bandwidth of DM designs. High negative dispersion $< -1000 \text{ fs}^2$ could be achieved only for bandwidths smaller than 70–80 nm. In Table 2 the most important parameters of obtained Nb₂O₅/SiO₂ designs are summarized.

4. Ta₂O₅/SiO₂ layer materials pair

Calculations similar to Nb₂O₅/SiO₂ layer material pair have been performed for Ta₂O₅/SiO₂ pair. In this case we also considered bandwidths of 20 nm, 40 nm, 60 nm, 100 nm, 200 nm and 400 nm. As in the previous case the termination condition was the total physical thickness of DM on levels of 10 μm , 15 μm and 20 μm .

Table 2. Summarized main parameters (the bandwidth, physical thickness, optical thickness, obtained value of GDD, number of layers) of designs with Nb₂O₅/SiO₂ and Ta₂O₅/SiO₂ layer materials pairs

$\Delta\lambda$, nm	Nb ₂ O ₅ /SiO ₂				Ta ₂ O ₅ /SiO ₂			
	Ph. th., nm	Opt. th., nm	GDD, fs ²	<i>N</i>	Ph. th., nm	Opt. th., nm	GDD, fs ²	<i>N</i>
20	9569.3	17189.2	−10000	87	9842.4	17715.6	−7000	64
20	14693.5	26472.9	−16000	125	14692.8	26445.6	−12500	78
20	20005.2	36620.0	−22000	167	19823.6	35682.5	−15000	107
40	9582.0	17114.4	−4000	91	9695.7	17278.3	−2200	89
40	15112.0	26998.7	−6000	133	14709.1	26505.3	−4000	123
40	21168.5	37773.3	−10000	181	19219.7	35266.1	−5500	144
60	10008.0	17825.6	−2000	84	9631.0	17265.9	−1500	85
60	15080.4	26900.0	−3000	120	14573.1	26113.9	−2500	119
60	20046.9	35964.2	−4500	167	19956.9	28500.4	−3000	129
100	9334.2	16704.9	−600	73	9741.4	17405.7	−700	86
100	15009.3	26564.9	−1000	132	14511.8	25898.5	−1000	117
100	17980.0	32508.7	−1500	148	19063.5	34003.0	−1500	157
200	10119.8	17913.7	−200	87	9686.1	17379.1	−250	91
200	15145.1	26633.2	−400	101	14100.3	25271.2	−400	128
200	20147.3	36537.3	−500	143	19780.6	35636.9	−550	151
400	11080.0	20054.8	−150	87	10097.3	18191.1	−100	102
400	15836.2	28633.2	−200	102	14845.2	26721.4	−150	134
400	19751.9	35750.5	−250	134	19923.6	35862.5	−200	158

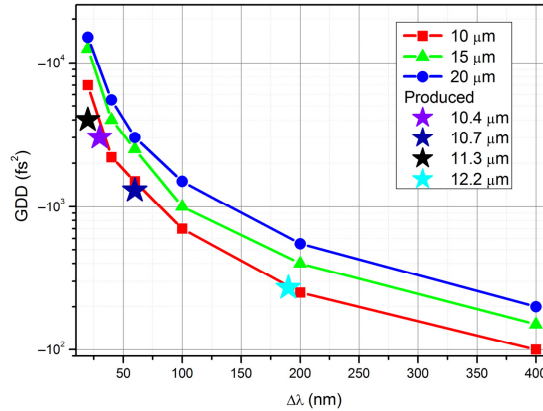


Fig. 2. Summarized main parameters of obtained designs with Ta₂O₅/SiO₂ layer materials pair. Red, green and blue curves correspond to designs with the maximum physical thickness of 10 μm, 15 μm and 20 μm, respectively. Asterisks correspond to produced designs [2,3,6,7] with physical thicknesses shown in the legend.

Figure 2 demonstrates that Ta₂O₅/SiO₂ layer material pair allows to obtain maximum negative GDD values that are generally smaller than for the case of Nb₂O₅/SiO₂ pair. This result is expected due to a higher refractive index contrast of the Nb₂O₅/SiO₂ layer material pair. It is also interesting to note that for large bandwidths (≥ 200 nm) maximum achievable GDD values are quite close. For the Nb₂O₅/SiO₂ layer material pair a dominating limiting factor for wideband DM designs is the short-wavelength absorptance in Nb₂O₅ layers. The most important parameters of obtained Ta₂O₅/SiO₂ designs are also summarized in Table 2.

5. Other layer materials pairs and TOD influence

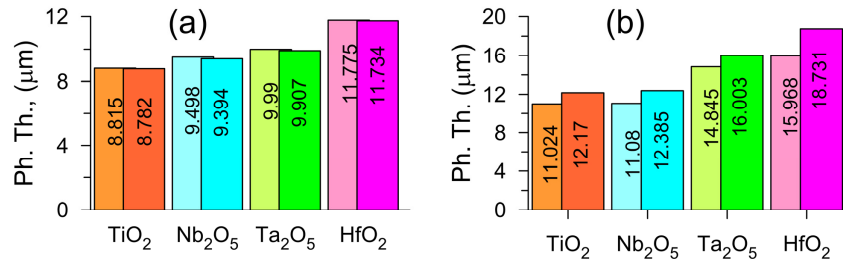


Fig. 3. Physical thicknesses of designs with different layer materials pairs (a) for 100 nm bandwidth (b) for 400 nm bandwidth: left bars correspond to TOD = 0 fs³ ((a) and (b)) and right bars correspond to TOD = -1000 fs³ (a) and to TOD = -150 fs³ (b).

The influence of a broader range of layer materials pairs is illustrated in Fig. 3. We considered the bandwidth 100 nm [Fig. 3(a)] and the bandwidth 400 nm [Fig. 3(b)]. Left bars in each material group correspond to a case TOD = 0 and right bars correspond to TOD = -1000 fs³ [Fig. 3(a)] and TOD = -150 fs³ [Fig. 3(b)]. These values of TOD provide slopes of GDD(λ) spectral dependence being larger than typical. TiO₂/SiO₂ material pair allows to obtain designs with somewhat smaller total physical thickness, than Nb₂O₅/SiO₂ pair in all cases due to higher contrast of refractive index values. HfO₂/SiO₂ pair requires noticeably higher total thicknesses and can be recommended for high intensity applications only. It is important to note that reasonable values of TOD can be easily taken into account almost without sacrificing the overall performance of the designs. Some increase of the total physical thickness can be noted only in the case of designs with 400 nm bandwidth.

6. Current state-of-the-art in DM production

Some of the obtained designs were experimentally produced. For the production we used magnetron-sputtering deposition plant (Leybold Optics HELIOS) with well-calibrated time control of layer thicknesses. Asterisks in Figs. 1 and 2 correspond to produced coatings. Most of realized designs have the total physical thickness about 10 μm or slightly more. Designs with the total physical thickness larger than 12 μm have not been produced yet due to high sensitivity of the design performance (especially GDD values) to manufacturing errors. Another potentially limiting factor is increase of stresses in a thick multilayer structure. The realization of such designs may be done in future with further improvement of layer deposition accuracy.

7. Conclusions

The main result of the current work is the empirical estimation of the maximum achievable negative values of GDD for different bandwidths and material combinations. Both layer materials combinations considered in detail demonstrate similar values of GDD, with slightly larger ones in the case of Nb₂O₅/SiO₂. These results give important practical estimations useful for designers of ultrafast laser systems. Further practical realization of DMs with physical thicknesses of 15 μm and 20 μm may provide increasing of maximum achievable value of GDD by the factor of 2.

Presented results were obtained by numerical computations and therefore there are opportunities to improve each design slightly. Nevertheless we believe that presented DM designs are quite close to fundamental limits.

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