

1.5 octave dispersive dielectric multilayers for pulse compression

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ABSTRACT

We demonstrate numerically and experimentally two types of dispersive mirrors with the controlled reflectivity and dispersion in a 1.5-octave bandwidth in wavelength ranges of 300-900 nm and 400-1200 nm. The measurement of the dispersion in a wavelength range of 300-1200 nm was performed. These types of dispersive mirrors allow one to correct the phases of the corresponding spectra, supporting sub-2-fs and sub-3-fs pulses, respectively.

Keywords: coatings, thin films, ultrafast optics.

1. INTRODUCTION

Generation of one-cycle pulses is a decade-long challenge in optics. The controlled superposition of light frequencies extending over more than one octave opens – along with carrier-envelope phase control [1-3]– the way to shaping the sub-cycle (i.e. sub-femtosecond) evolution of light fields in laser pulses and thereby to a new way of quantum control based on light-force directed charge in atoms, molecules or solids [4]. There are two main approaches to generate one-cycle pulses: i) synthesis of different laser sources [5-7] or Raman generator based on synthesis of vibrational or rotational transitions in molecules [8-9] with its further compression by using spatial light modulator (SLM) [8], and ii) compression of supercontinuum behind hollow gas-filled fiber [10-12]. Further progress in these two approaches is limited by both low transmission of SLM and the highest pulse energy that can be applied to SLM.

An alternative to SLM can be a broadband dispersive (chirped) mirror (DM) compressor. DMs have been continuously improved since their invention in 1994 [13]. Progress has been made in terms of bandwidth, losses, group delay dispersion, and the ability to compensate higher-order spectral phase errors introduced by optical components [14-22]. As a result of the efforts of several groups, by the turn of the millennium DM-based optical systems have been capable of controlling broadband radiation over spectral ranges approaching an octave in the visible-near-infrared domain [7,10, 11, 23, 24].

2. DESIGNING OF A DM

A DM is a dispersive optical interference coating usually designed by optimizing the initial multilayer design. A DM is characterized by a certain value of the group delay dispersion (GDD), the second derivative of the phase shift on reflection with respect to the angular frequency. A DM can provide a broadband spectrum with support, comparable with prism and grating pairs, but additionally it offers control of third and higher-order dispersions and higher efficiency (reflectivity) together with better beam stability. Reflection from the top layer of a multilayer structure brings so-called ripples to the spectral GDD curve due to interference between waves reflected from the top layer and waves which have penetrated and been reflected from deeper layers. Ripples become pronounced in the case where air is an incident medium. In general, the mirror GDD should compensate material (through which the initially short pulse passes) or the (nonlinear) pulse chirp so that the residual dispersion fluctuations are acceptably small in all of the relevant spectral range. Usually, during design optimization, residual fluctuations drop to a low level. The GDD fluctuations can broaden the pulse and lead to energy transfer from the initial single pulse to satellites. The period of the ripples in the spectral domain determines the position of the satellite in the temporal domain, and the amplitude of these oscillations determines the amount of energy which transfers into the satellite (s).

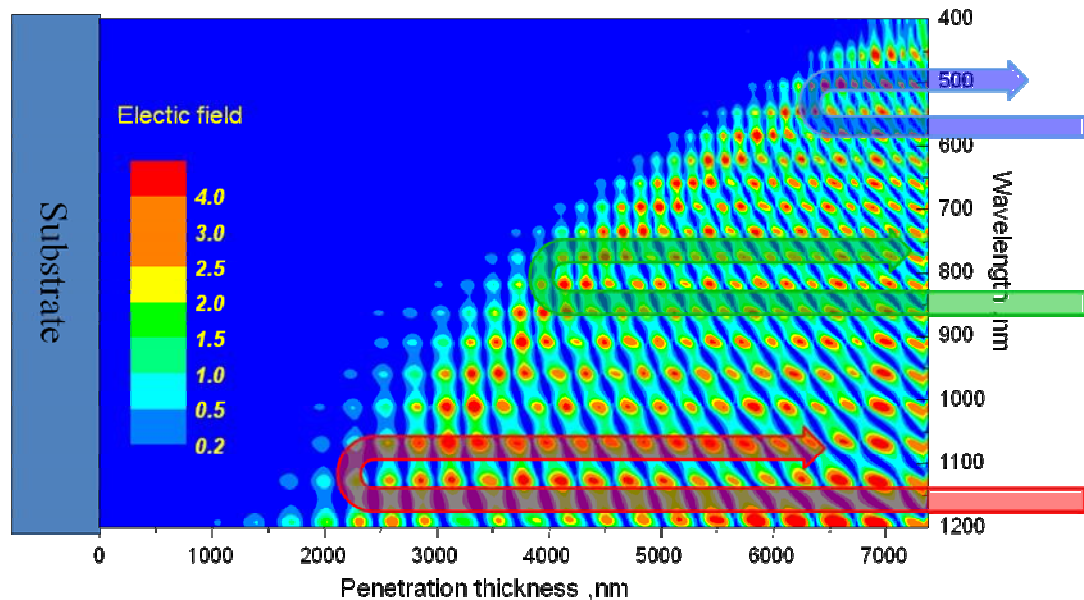


Fig. 1. The penetration depth of different spectral components into the structure. The electrical field of longer wavelengths penetrates much deeper into the multilayer structure than the shorter wavelengths. This means that spectral longer components become delayed relatively to shorter components.

A bandwidth of a DM and its GDD spectral shape are defined by, among other parameters, impedance mismatch between the ambient medium and mirror stack. The impedance mismatch can be overcome by using glass as medium of incidence. Unfortunately, this solution limits the highest value of GDD, because the mirror stack should compensate an additional thin unparallel substrate [15] or glass wedge [16]. Additionally, in this case the aperture cannot be high and the beam stability problem may arise. There are several other approaches to reducing GDD ripples: double-chirped mirrors [14], Brewster-angled DM [18], complementary CM pairs [20]. Alternatively, time-domain optimization [19, 23] deals directly with pulse compression and can therefore lead to the shortest pulses even in spite of large GDD oscillations. The complementary mirror approach has high potential and the relevant results are shown below.

For providing pulse compression down to sub-3 fs, it is necessary to design a complementary DM pair with smooth dispersion and high reflectivity in all of a 1.5-octave spanning spectrum. Note that the requirement of dispersion smoothness is greater for a broader spectrum. An appropriate design of such an ultrabroad DM (UBDM) is a complex thin-film problem requiring fast optimization algorithms and a powerful computer to calculate a large number of target points.

In this work the OptiLayer software was used for designing the mirror. The distinguishing feature of OptiLayer is its completely analytical approach to computing all quantities necessary for coating optimization [25]. This feature provides a computational efficiency that cannot be achieved when non-analytical approaches are applied to coating optimization. Note that the characteristic GDD to be optimized is the second derivative of the phase shift with respect to the angular frequency, and modern high-efficient optimization methods also require second derivatives of this characteristic with respect to layer thicknesses. Utilizing the completely analytical approach to computing all derivatives required provides fast computation and high accuracy of the synthesis algorithms at all stages of designing.

The first pair of UBDM consists of 78 and 80 layers. This pair provides a negative GDD -20 fs^2 at 800 nm. The mirror design was chosen to compensate the dispersion of 3 m of air and 3 mm of fused silica for 10 bounces. The reflectivity and the GDD curves of this pair are shown in Fig. 2. For the UBDM pair we avoided the inconsistency in the periods, which was present for UBCM. Sharp thin spikes in both the reflection and the GDD curves are clearly seen in the range 400-450 nm in Fig. 2. The appearance of sharp spikes is typical of ultrabroadband reflectors but theoretical discussion thereof would exceed the scope of this publication. Suffice it to mention here that this phenomenon is associated with resonance effects inside a high-reflection multilayer structure at certain wavelengths [26]. The typical width of the GDD spikes for the UBDM pair is smaller than 0.3 nm. We found numerically that these spikes do not lead to satellites in the time domain, contrary to ripples.

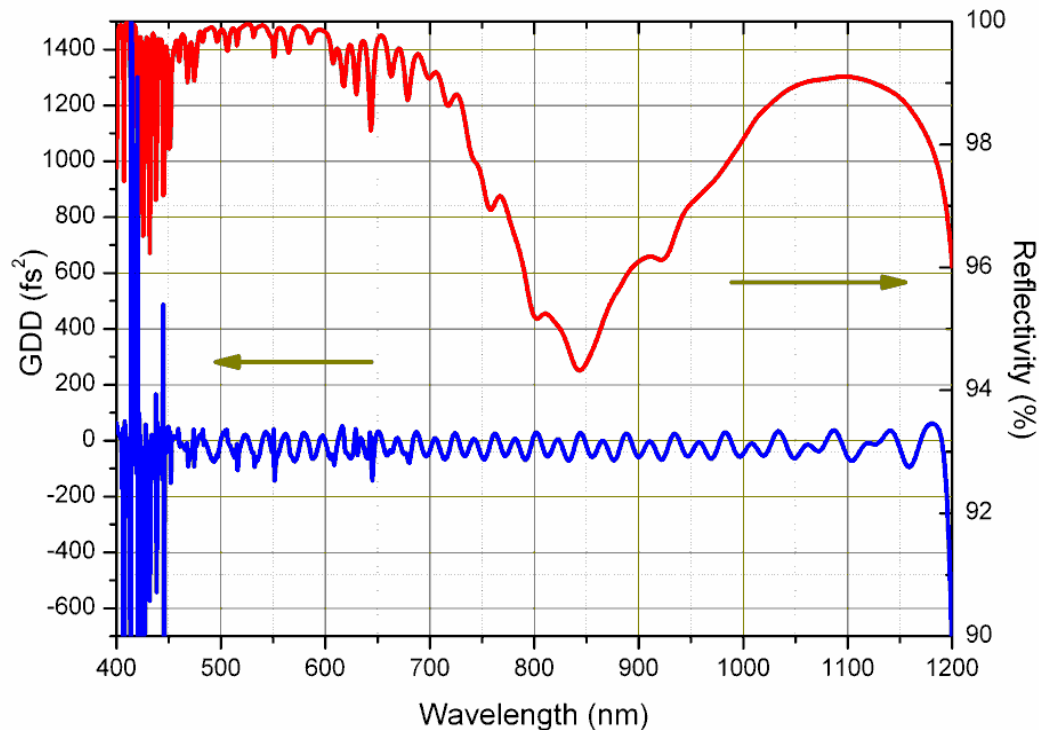


Fig. 2. Calculated reflectivity (red line) and GDD (blue line) averaged for a complementary pair of DMs.

Figure 3 shows results of temporal pulse analysis of the pulse reflected after several bounces of the UBDM pair. In the analysis the main part of the dispersion was taken away and only residual GDD ripples were included. We used a Gaussian free-chirp incident pulse centered at 800 nm (spectrum 400-1200 nm) with the spectrum corresponding to a bandwidth-limited pulse (FWHM) 2.48 fs. The reflected pulse does not become longer when the ripples are small or absent. Calculations for the UBDM pair demonstrate that after 10 bounces i) the incident pulse extends only up to 2.64 fs and ii) the energy losses of the pulse are 50%. This value can be considered as acceptable at this initial stage of research and bearing in mind ~15% of the SLM throughput [27].

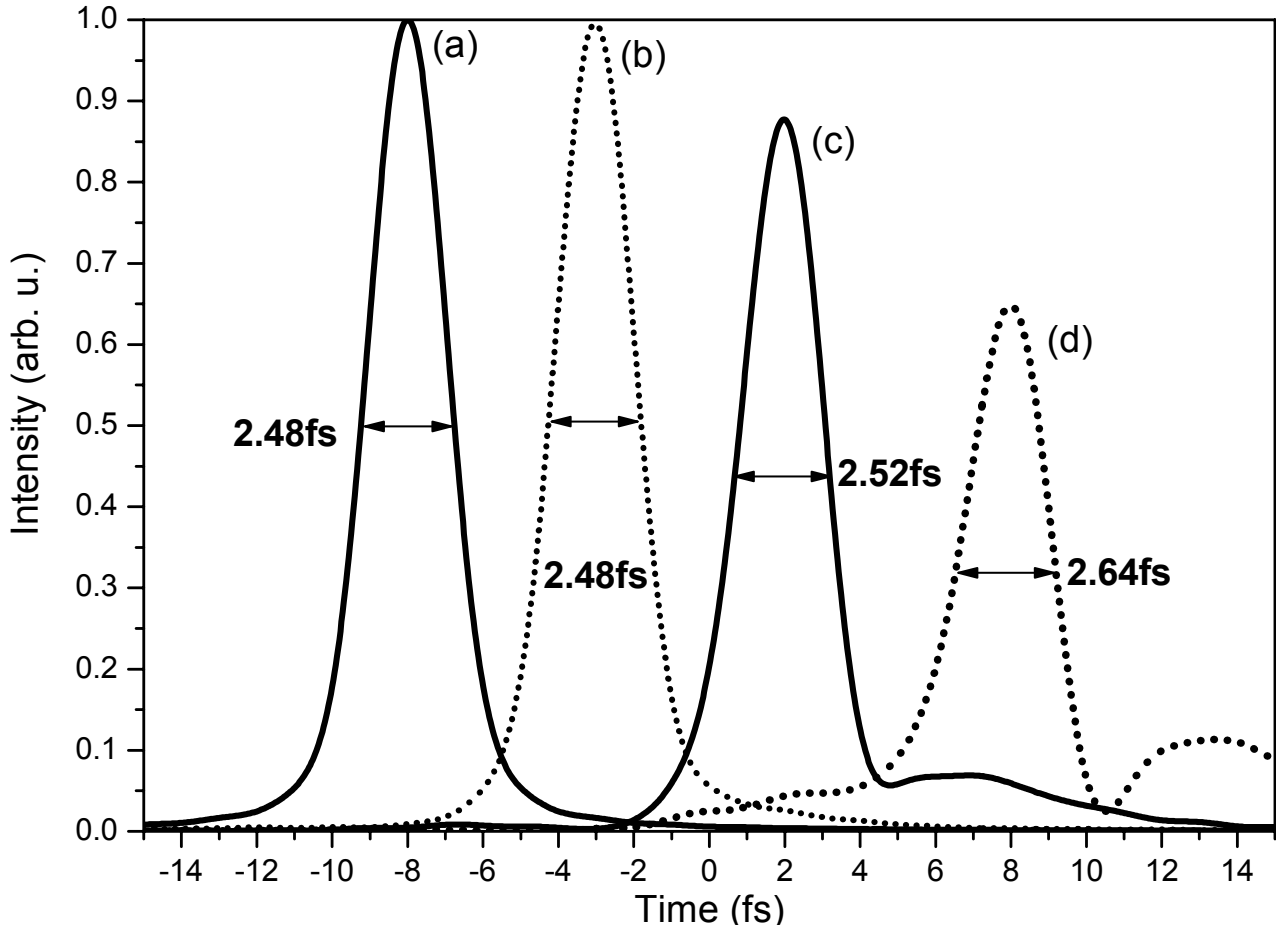


Fig. 3. Temporal analysis of the reflection of an incident Gaussian-shaped free-chirp 2.48-fs pulse (a); (b) after one bounce from the UBDM pair; (c) – after 5 bounces from the UBDM pair, and (d) – after 10 bounces from UBDM. The temporal shift of the curves is artificial.

The second UBDM pair HfO₂/SiO₂ was designed for a spectral range of 300-900 nm. The refractive index of hafnium oxide ($n_H \sim 2.05$ @ $\lambda \sim 500$ nm) is much smaller than that of high-index materials, such as Nb₂O₅ ($n_H \sim 2.35$) and TiO₂ ($n_H \sim 2.45$). Achieving a high reflectivity over a large bandwidth with such a low value of n_H is a formidable challenge for our HfO₂/SiO₂ UBDM. Our resulting design consists of 83 alternating layers of HfO₂ and SiO₂ and exhibits a spectrally-averaged reflectivity of 93 % in the 300-900 nm spectral range and a nominal group delay dispersion of -20 fs² at the center wavelength of ~ 450 nm [28, 29]. To analyze the potential the UBDM HfO₂/SiO₂ mirrors offer for pulse compression and optical waveform synthesis, we have considered a hypothetical broadband dispersive delay line with 5 bounces off each of the two complementary dispersive mirrors (total GDD at 450 nm ~ -200 fs²) compensating the dispersion of 2 mm fused silica and 0.4 m of air. The measured GDD curve over the 300-900 nm range was supplemented with the design curve. We have sent a hypothetical Gaussian pulse carried at a wavelength of 450 nm 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 mentioned above. The results are summarized in Fig. 4. The uncompensated

spectral oscillations in the mirror GDD curve [28] lead to a broadening of the pulse to approximately 2.2 fs, accompanied by a decrease of the pulse energy by a factor of ~ 2 .

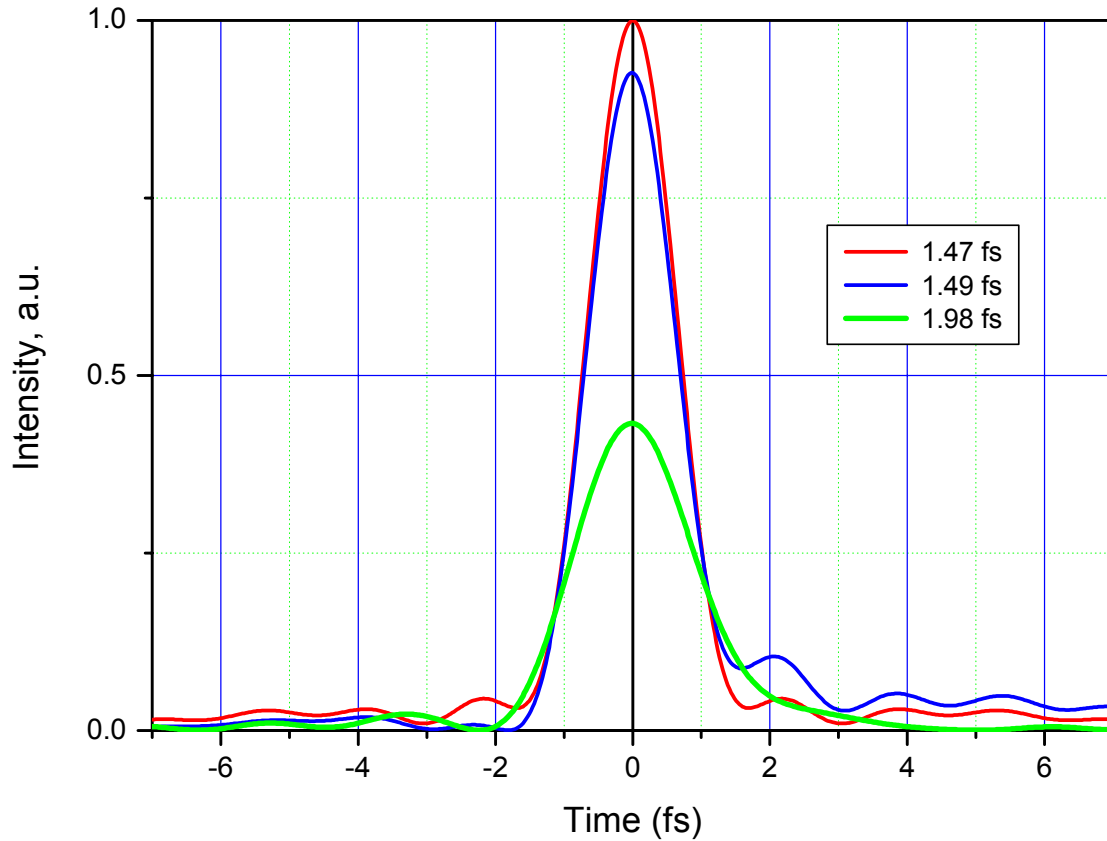


Fig. 4. Evolution of an incident pulse (red) after 1 bounce (blue) and 10 bounces (green).

3. DISCUSSION

The DMs were manufactured by Helios Leybold Optics sputtering system. Important to note that the refractive indexes of Nb_2O_5 ($n=2.35$) and SiO_2 ($n=1.48$) is higher than those achievable with electron beam evaporation, indicating very dense layers and broader spectral ranges of mirrors. Ultimate precision in layer growth control is facilitated by an optical monitoring system for in-situ measurements. The highly stable middle frequency magnetron sputtering process allows precise layer thickness deposition by time control, in cases where no optical monitoring is used. Spectral measurements of both UBDM show very good agreement with the design in respect of both reflectivity and dispersion, see comparison of Fig. 2 and 5. If the calculated and measured GDD curves are close to each other, the errors made during deposition are small or it is the result of effective error compensation. Compensation means here that a smaller refractive index can be compensated by thicker physical thickness of the layer, thus keeping the optical thickness constant. A good agreement between the design and measurements in the range 400-1100 nm led us to the assumption that the GDD curve behaves in a predictable way in the range 1100-1200 nm as well.

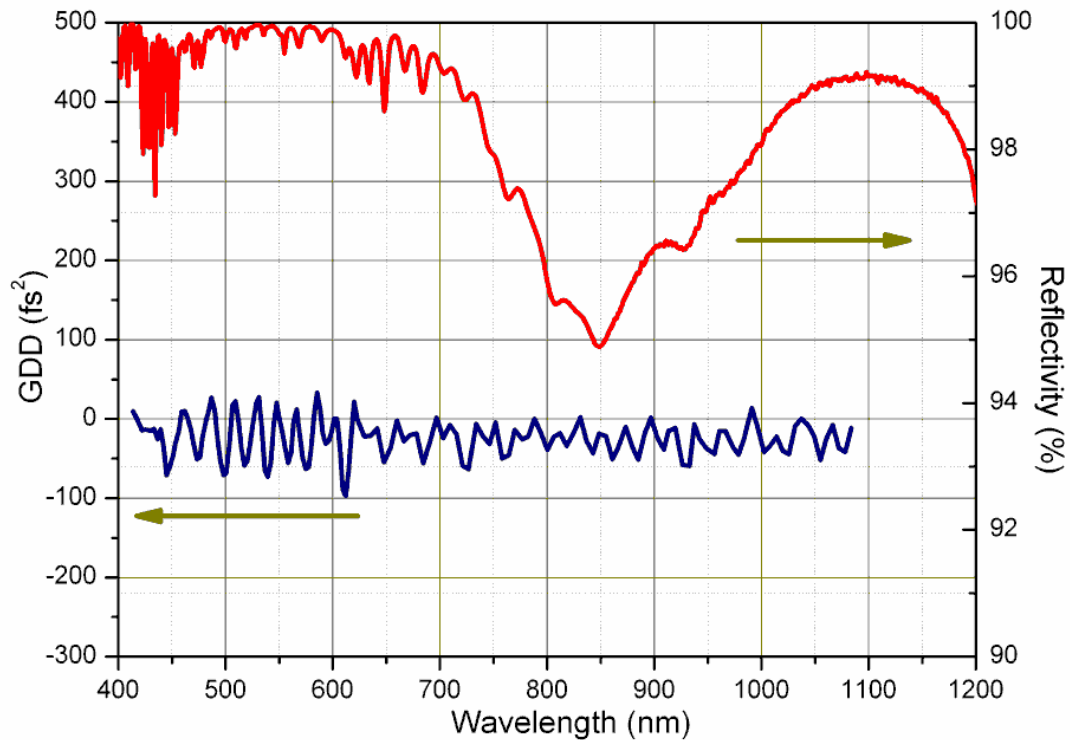


Fig. 5. Measured reflectivity (red line) and GDD (blue line) averaged for a complementary pair of DMs.

The reflection spectra were measured with spectrophotometer PerkinElmer (Lambda 950). GDD was determined by using white light interferometer with the spectral accuracy 10 nm and dispersion accuracy $\sim 10 \text{ fs}^2$. One-mirror measurement together with the data retrieval takes $\sim 0.5 \text{ h}$.

4. CONCLUSION AND FUTURE VISION

We have reported the extension of ultrabroad-band dispersion control with dispersive dielectric multilayers well into the ultraviolet, visible and infra-red spectral ranges. The reported complementary pairs of dispersive multilayers offer the potential for mono-cycle pulse generation in the blue-infrared spectral range and lends itself to applications in future ultrabroadband optical waveform and frequency-comb synthesizers. Thanks to the high damage threshold of DM, this work also opens the way to the development of dispersive multilayer for applications in high-power laser systems.

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