Efficient broadband highly dispersive HfO_2/SiO_2 multilayer mirror for pulse compression in near ultraviolet

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Abstract: We report on design, production and implementation of a highly dispersive broadband dielectric multilayer mirror covering near ultraviolet range from 290 nm to 350 nm. The described mirrors, having 92% spectrally averaged reflectance in the ultraviolet range and $\sim$ 85 fs of group delay difference, that allow compression to $\sim$ 7 fs, provide a strong foundation for generation of few-fs pulses in the near ultraviolet.

OCIS codes: (320.5520) Pulse compression; (310.1620) Interference coatings; (310.6845) Thin film devices and applications.

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1. Introduction

Investigation of fast evolving dynamics of photo active biological and chemical systems via exploitation of time-resolved spectroscopy [1] requires few-cycle pulses with central wavelengths in the range from near ultraviolet (UV) to vacuum UV (VUV). Many advances in the development of such sources have been made in last decade [2–8]. Despite the great efforts, generation of high-energy few-fs UV pulses remains non trivial.

Synthesis of few-cycle pulses in UV is acknowledged to be more difficult, due to the significant refractive index dispersion in the UV of the most commonly used optical materials, including gases, that causes temporal broadening of the synthesized pulses. Except for the direct frequency upconversion in a gas jet, which yields nearly transform-limited pulse [4, 5, 9], most other methods require post compression. There are several approaches for dispersion control in ultrafast laser systems, which are valid not only for visible (VIS) and near infrared (NIR), but also to some extent for UV. Grating compressors offer substantial dispersion management possibilities, although exhibit relatively low transmission in UV ($\leq 50\%$ [3]). Using of prism preserves best efficiency (up to 93% [10]). However, achieving sub-10 fs pulses requires special treatment to avoid spatial- and mode- distortion [11] and precise prism matching to guarantee compensation of the third order dispersion. Conventional pulse shapers [12] applicable to UV include micro mirrors (MEMS), deformable mirrors [13] or acousto-optic programmable dispersive filters (AOPDF) [14]. However, deformable mirrors and MEMS are limited by the maximum amplitude of the deformation [13], while AOPDF is doomed by rather low efficiency in UV (about 20% for 30 fs pulse in case of AOPDF [10]), though all provide unprecedented flexibility for pulse shaping.

Well known alternative approach for post compression of ultrafast pulses is implementation

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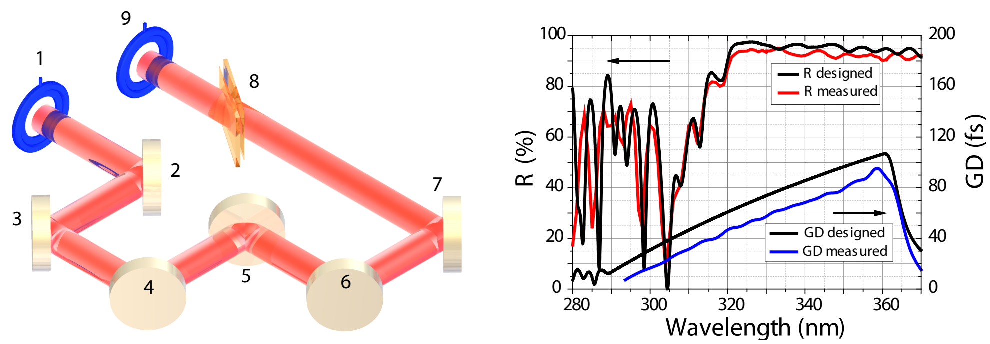
of dispersive dielectric multilayer mirrors (DM), also referenced as chirped mirrors (CM) [15]. Apart from their high efficiency and possibility to operate on super broadband spectra [16], DM-based compressors are very attractive due to the simplicity of the set up. Unlike in most pulse shapers [12], neither spatial chirping, nor fixed spatial separation are required to achieve specified dispersion. Therefore, DM-based compressors are robust, user friendly, and can be very compact. The concept has been successfully explored in VIS and IR spectral regions, expanded to VUV and extreme UV (EUV) [17, 18] and approached the range of near UV [19]. However, overall the UV spectral region still remains poorly covered [20, 21], partially due to the lack of suitable coating materials that would have low absorbance in UV.

Here we present broadband highly dispersive mirrors designed for the central wavelength of 325 nm, that were successfully implemented for post compression of the UV pulse down to ~7 fs [22]. By tuning the structure of the multilayer stack, we managed to significantly suppress the linear and nonlinear absorbance [23] and reach the overall reflectance of 94% and the total compressor efficiency of $\geq$ 50% in straight forward set up. Further development promises continuous improvement to the compressor's efficiency.

2. Design and production

The compressor was designed to compensate for material dispersion accumulated by few-fs pulse after propagation through 3.5 mm of magnesium fluoride and 4 m of air path in the spectral region from 290 nm to 350 nm. Angle of incidence (AOI) is 45 deg, incident light is p-polarized. The compensation is to be achieved with six bounces on UV DMs, Fig. 1(a). Designed GD per reflection, fulfilling the requirements, is presented in Fig. 1(b), designed reflectance at 45 deg is presented in Fig. 2(c).



(a) (b)

Fig. 1. UV DMs compressor. (a) Compressor set up. 1,9 - irises, 2-7 - DMs, 8 - wedges. (b) Design and production. The solid black curves represent design data, the solid red, blue curves - data measured after production. The reflectance is shown for AOI = 7 deg, p-polarized light, GD - for AOI = 45 deg, p-polarized light. The vertical offset of GD curves is introduced for illustration purposes.

Design of the DMs for UV requires special considerations. The spectral ranges, that can be served by DMs are defined by the transparency regions of used coating materials. As the evolution of DMs started together with evolvement of Ti:Sapphire based systems, the most effort was put into development of technological process for materials covering VIS and NIR, that are, regrettably, non-transparent in UV. Despite that recently HfO_2/SiO_2 material pair, appropriate for the UV range, has started being used for production of dispersive coatings in UV [25],

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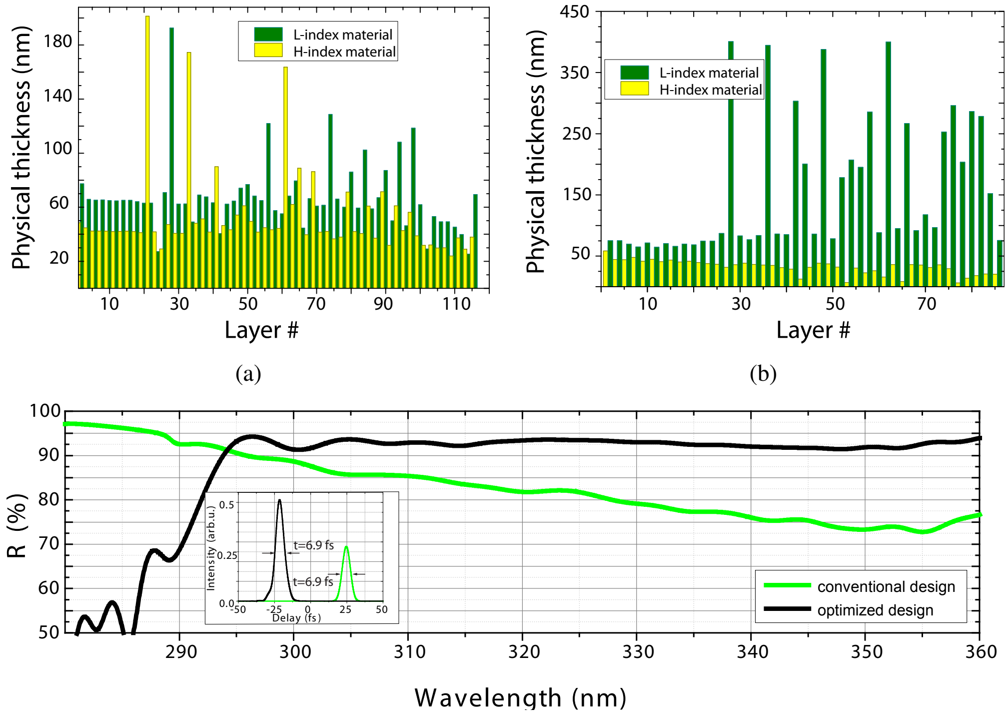
the achievable reflectance is still limited by linear [24] and to some extent nonlinear [23] absorptance of the materials of the coating.



(a) (b) (c)

Fig. 2. Optimization of the UV DM. (a),(b) - layer sequences of conventional (a) and optimized (b) mirror designs. (c) - theoretical reflectance at 45 deg AOI, p-pol light of both designs together with simulated pulse duration achieved after six reflections from described DMs (inset). Both mirror designs support 6.9 fs pulse.

In our recent work [23] we have demonstrated how by tuning the structure of the stack, one can significantly suppress both the linear and the nonlinear absorptance. We have followed this approach in the design procedure of the UV DM. Figure 2 represents two designs of the UV DM. The conventional design was developed without taking minimization of the absorptance into consideration, while optimized design accounts for linear and nonlinear absorptance. Tuning of the UV DM design allowed to reach higher reflectance in comparison to conventional approach, Fig. 2(c). Spectrally averaged reflectance of the modified design is 94%. The difference between the designs is clearly seen. The optimized design, Fig. 2(b), not only contains less of high-index material (1.38 μm against 3.07 μm for conventional design), but the material is distributed differently. In the optimized stack the high-index material is redistributed farther from the upper layers, where electric field becomes weaker, unlike in case of conventional design, Fig. 2(a), where distribution is homogeneous. The total thickness of the designs is also different (6.98 μm for conventional design, 8.14 μm - optimized). In order to suppress the absorption, the significant part of the high-index material is substituted with the low-index counterpart. Meanwhile, in order to preserve the GD, particular minimal optical thickness is needed. Therefore, the physical thickness of the low-index layers needs to be increased and, in consequence, the overall thickness of the optimized designs is typically higher, while the

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amount of layers is less.

Mirrors were coated on fused silica substrates via magnetron sputtering deposition technique (Helios plant, Leybold Optics). Reflectance data were measured in spectrophotometer (Lambda-950, Perkin Elmer). The design of the machinery allows reflectance measurement at 7 deg AOI only, therefore in Fig. 1(b) the designed and measured reflectance curves are presented at 7 deg AOI. GD was measured in the home-made UV white light interferometer, that covers spectral range from 250 nm to 460 nm. Spectral resolution of the spectrometer, implemented as a detector, is 1 nm. Total amount of 30 data points with a spacing of 2.5 nm was used to fit the measured phase data and extract the GD. The spacing is chosen basing on the period of an anticipated GD ripples. The dispersion characteristics were measured at 45 deg AOI in p-polarized light. Acquired reflectance and GD data are presented on Fig. 1(b). Comparison of designed and measured GD shows convincing correspondence. The measured reflectance is somewhat lower, the spectrally averaged reflectance is 92% versus 94% in the design. This discrepancy can indicate either imperfection of the deposition process and scattering losses, or slight imprecision in the estimation of the induced losses. In latter case, additional tuning of the design will allow to further improve the performance.

3. Implementation

Before implementation, we have performed temporal pulse analysis in order to evaluate potential performance. For our calculations we used a Gaussian chirp-free pulse centered at 325 nm with a spectrum spanning from 290 nm to 360 nm, which supports 5.8 fs Fourier-transform-limited (FTL) pulse (black solid curve Fig. 3). Six bounces on the UV DM yield 6.9 fs pulse containing approximately 50% of incident intensity (blue solid curve Fig. 3). The simulation was found to be encouraging, as we have reached efficiency much higher than the one reachable with AOPDF and comparable to gratings compressors in a set up, that is much simpler than prism arrangement.

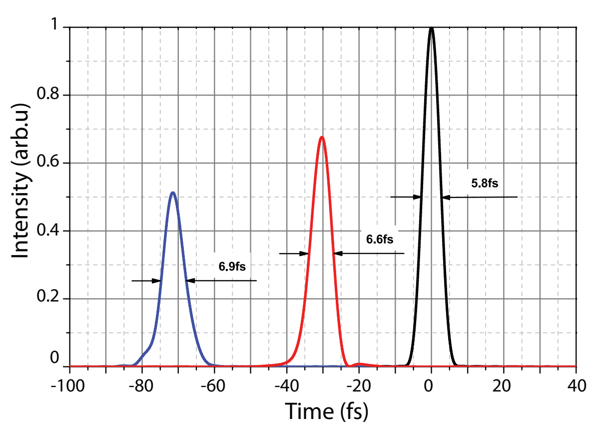

Fig. 3. Pulse analysis. Simulated performance of the compressor set up. Black solid curve incident FTL pulse, blue solid curve - pulse after six bounces on described UV DM, red solid curve - prognosed yield of next generation UV DM.

The UV DM were build into the compressor set up. A pair of thin magnesium fluoride wedges was used in addition to six UV DM for the fine tuning of the dispersion. At first attempt, the achieved pulse duration did not reach the predicted 6.9 fs. After reconstruction of obtained

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FROG traces it was concluded that spectral phase of chirped pulse includes not only material dispersion, compensated by the mirrors, but has an influence of higher orders dispersion inherited from the parental nonlinear process. The design target had to be corrected in accordance. A customized set of UV DMs was produced. Their implementation allowed post compression down to 6.5 fs [22] corresponding well to the theoretical estimation of 6.9 fs, shown in Fig. 3, for initial design. The total throughput of the system reached 53%, which is also well within the estimation.

4. Conclusion and outlook

In our work we have demonstrated the implementation of the UV DMs for compression of broadband pulses in near UV. The mirrors were carefully designed taking into consideration linear and nonlinear absorptance of the coating materials. As a result, the spectrally averaged reflectance of the DMs of 92%, overall efficiency of the compressor of $\geq 50\%$ and compression down to 6.5 fs were demonstrated.

Targeting the improvement of the compressors' performance, we have looked into upgrading of the UV DM design. Further tuning of the multilayer stack by substituting the high-index material in the stack with low-index material will allow to increase the compressor's efficiency to $\geq 65\%$ (red solid curve in Fig. 3). If reachable, this efficiency will outreach the gratings set ups. However, it will come at expense of increase in total thickness and possible appearance of tensile stress, that affects mechanical properties of the coating.

It's worth remarking that, due to the limitation of the current set up to 45 deg AOI and p-pol light, the reachable theoretical reflectance is a priori lower than for the other cases. Designing the DMs for small AOIs or s-pol light will additionally improve achievable reflectance.

Further expansion of the dispersive mirror technology into the UV in principle calls for the implementation of the different material pair. From the perspective of the minimization of the absorptance, the Al_2O_3/SiO_2 material pair is the most proper choice. Regrettably, the contrast between the refractive indices in case of Al_2O_3/SiO_2 (1.79/1.47 at 365 nm) is much smaller than in case of HfO_2/SiO_2 pair (2.18/1.47 at 365 nm) [26]. In consequence, in order to reach the comparable spectral bandwidths, high GD and reflectance, more layers need to be deposited. As the deposition error scales proportionally to the number of layers and inverse proportionally to the layer's thickness, the production of the high precision coatings for UV from Al_2O_3/SiO_2 will become increasingly challenging. The required precision is reachable with implementation of the ion-beam sputtering deposition technology.

Overall, simplicity and compactness of mirror-based compressors, along with good efficiency and manifold compression to nearly FTL pulse, make them a promising perspective in application to the few-cycle pulses in UV/near UV ranges.

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