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Review Article

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Dispersive mirror technology for ultrafast lasers in the range 220–4500 nm

Abstract: Nowadays, dispersive mirrors are able to cover the wavelength range of 4.5 optical octaves and can be used from 220 nm up to 4500 nm. Various design approaches to dispersive mirrors in visible and near IR are briefly discussed. We consider in more detail two dispersive mirrors representing extreme cases. The first one is a mirror working in the range of 290–360 nm and providing group delay dispersion of -75 fs^2. The second one is a mirror working in the range of 2500–4500 nm and providing +500 fs^2 of group delay dispersion.

Keywords: chirped mirrors; dispersive mirrors; interference coating; ultrafast coatings; ultrafast lasers.

OCIS Codes: (310.4165) multilayer design; (320.5520) pulse compression; (310.6805) theory and design.

1 Introduction

Dispersive mirrors have played a pivotal role in expanding the frontiers of ultrafast technology since their invention in 1993 [1]. They allowed pulse duration to approach the period of a carrier wave and, more recently, shaping of the waveform within the wave cycle and the synthesis of subcycle light transients [2–4].

In the 1990s, the term ‘chirped mirrors’ and its modifications (e.g., ‘double-chirped mirrors’) were widely used in the field of multilayer coatings aimed at ultrafast applications. Typically, chirp mirrors were designed as multilayer stacks with layer thicknesses decreasing in the direction from substrate to incident medium according to some predefined law. The working principle of chirp mirrors was based on penetration depth effect, i.e., on the reflection of different frequencies of the pulse spectrum from layers positioned at different depths. Later on, the term ‘dispersive mirrors’ become more appropriate due to utilization of resonant effects (Gires-Tournois resonance cavities) and complicated combinations of resonance and penetration depth effects. Nowadays, complicated dispersive mirrors are designed with the help of powerful numerical algorithms and may have many dozens of layers.

During the last 20 years, dispersive mirror (DM) technology [1, 5–17] has become very popular in femto- and attosecond physics [4, 3]. Nowadays, the majority of femtosecond lasers include DM optics, which provides an accurate group delay (GD) or group delay dispersion (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 duration of <2 fs in the visible range [4]. Therefore, the design of DMs with required spectral performance (reflectance and GD/GDD characteristics) is crucial for many modern applications in the field of ultrafast physics.

Multilayer design and deposition techniques had remarkable development during the last two decades. As a result, it is possible to reliably produce multilayer coatings consisting of many dozens and even hundreds of layers. Most of the success was achieved in the UV, Visible, near- and far-IR ranges, yet EUV and X-ray multilayer coatings were also developed and produced successfully [18, 19]. It is necessary to mention that the wavelength range of 60–200 nm is hardly possible for high-reflective,

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low-loss multilayer optics due to the lack of suitable layer-forming materials for this range. For wavelengths below 60 nm (EUV range 10–60 nm, and soft X-ray range 0.1–10 nm), multilayer coatings are available, and even DMs have been produced [20–23]. These wavelength ranges have many specific features and peculiarities deserving a separate review, and we will not consider them here.

In this review, we discuss the tremendous progress in dispersive mirrors covering different spectral ranges from 220 nm to 4500 nm. These DMs possess unique dispersive properties (see Figure 1) [5–23, 27–34]. This progress opens a new perspective to the synthesis of arbitrary light fields [2, 4] and to the exploration of electron/atom interaction [3, 35] on a new level of knowledge, as well as to the evolution of tools for biological, chemical, and ecological studies.

2 DMs span the 220–4500 nm wavelength range

The generation of ultrashort pulses demands a laser beam with broad electromagnetic spectrum, the components of which have to be phase matched with each other in order to form and maintain the shape of the pulse and its duration. This, in turn, means that the spectral phase of the pulse has to be controlled, and the dispersion of the materials, through which the pulse propagates, has to be compensated. In the last 5 years, a broad family of UV, Vis, and IR DMs covering the spectral range from 220 nm to 4500 nm and possessing unique dispersive properties has been developed (see Figure 1).

Several of the mirrors have bandwidths of about one optical octave. In their working range, the mirrors provide high reflectivity and controllable phase. It is important to note that reflectivity degrades to shorter wavelength, which is to be expected due to the increased absorption in UV for most of the materials employed for DM designs. Nowadays, there are many commercially available DMs working between 500 nm and 1100 nm. On the other hand, there are almost no commercially available DMs for UV and IR ranges. In the next sections, we will demonstrate the ability of DMs to work in the range covering wavelengths from 220 to 4500 nm. The most recent advances in DM technology can be fully illustrated by two extreme cases: i) broadband DM in UV for the range 290–360 nm and ii) super-broadband IR DM for the range 2500–4200 nm.

3 Design approaches of DM in the Vis near-IR ranges

The main problem of DM design is related to unavoidable GDD oscillations, which appear due to interface mismatch

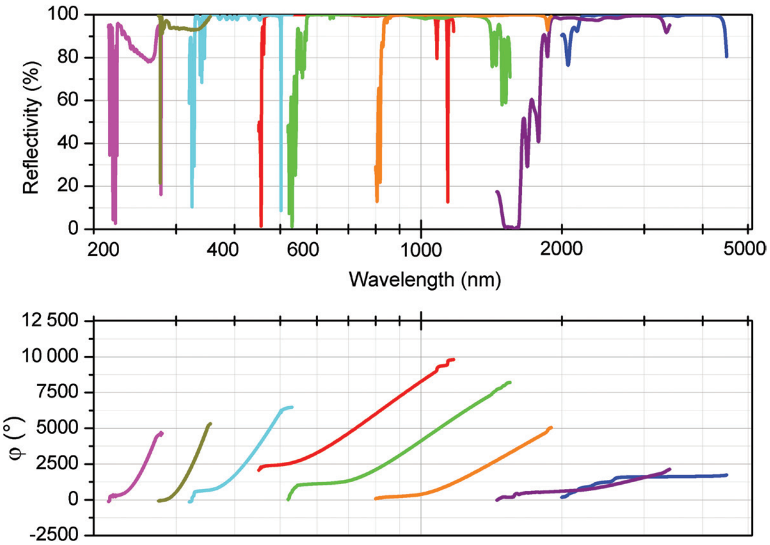

Figure 1 Spectral region covered by designed and produced dispersive mirrors. Reflectivity (upper box) and phase (lower box) are shown. Magenta curves correspond to DMs with a working range of 220–290 nm. Dark yellow curves correspond to DM described in Section 4. Cyan – DM with a working range of 350–500 nm [4]. Red – DM with a working range of 500–1050 nm [13, 24]. Green – DM with a working range of 700–1350 nm [25]. Orange – DM with a working range of 1000–1800 nm [http://www.ultrafast-innovations.de.]. Violet – DM with a working range of 1900–3200 nm, described in Section 5. Blue – DM with a working range of 2500–4200 nm [26].

between the top layer of the multilayer structure and the external medium. These irregular GDD oscillations lead to destruction of the pulse shape. This problem is especially important for broadband (about one optical octave) DMs. Several approaches have been devised for suppressing or overcoming these undesirable oscillations:

1. Double-chirped mirrors – the multilayer Bragg structures, which can be decomposed into a series of symmetric index steps, as shown in [31, 36]. The mentioned structure with an antireflection (AR) coating on the top can provide smooth GDD characteristics allowing the compression of the pulse down to a duration of 6.5 fs. AR makes it possible to compensate the interface mismatch between the top layer and the external medium, resulting in GDD characteristics with much lower oscillations. The main limitation of this approach is the relatively narrow operating bandwidth. Double-chirped mirrors cannot be used for bandwidth wider than half of an optical octave.
2. Resonant structures or Gires-Tournois interferometer (GTI) mirrors [5, 37] – two high-reflecting structures separated by a cavity with thickness close to half wavelength of the incident radiation resonantly enclose the impinging wave. Such nanoscale GTI embedded in the multilayer structure can introduce large GDs at selected wavelengths. The GTI approach is realized by optimizing the last few layers in symmetrical quarter-wave layer stacks that form the reflecting mirrors. A standard quarter-wave stack has zero GDD at the center of its stop band with small variation over the entire high-reflectivity zone. As a result of additional optimization, the required smooth GDD characteristics can be obtained, but the performance with a single GTI is satisfactory only in a narrow spectral zone. Using this approach, it is difficult to design DMs with an octave bandwidth because it requires too many resonant cavities.
3. Brewster-angle DMs – the elegant way of avoiding the cause of oscillations due to interface mismatch. This approach is based on using p-polarized light at the Brewster angle [34]. Placing a DM at the Brewster angle for the top layer ensures only a small amount of Fresnel-reflected light, resulting in almost oscillation-free GDD. Unfortunately, this approach requires almost twice as many layers as a DM at normal incidence to obtain the same reflectivity. Additionally, it is unrealistic to use the Brewster-angle DM as a laser oscillator mirror because the intracavity beam bounces at a small incidence angle.
4. Backside-coated DMs – the backside-coated DMs were proposed by N. Matuschek et al. in [38]. The authors suggest using one-side wedged substrate. The radiation penetrates through the wedged side of the substrate and interacts with the multilayer structure. The wedge splits spatially the Fresnel reflection from the top layer and the main part of the irradiation reflected from the inner layers. Such a design can be used to obtain a smooth GDD characteristic. Unfortunately, the substrate has to be a few millimeters thick to stay flat with a relatively thick dielectric multilayer on the top. The positive GDD accumulated by the beam during double pass through the substrate significantly diminishes the resulting GDD performance of the backside-coated DMs.
5. Tilted-front-interface DM – to avoid Fresnel reflection from the top layer, the wedge is now attached onto the top layer [32]. The tilted-front-interface DM requires an additional anti-reflection (AR) coating on top of the wedge to minimize losses. The requirements for AR of the coating on top of the tilted-front-interface DM are not as strict as for double DM. The Fresnel reflection of light from the wedged top brings only additional losses, but it has no influence on the GDD oscillations. The weakest point in the tilted-front-interface DM approach is connected with technological problems for applying a wedge onto the top of the multilayer stack. Until now, only a small-diameter (half inch) tilted-front-interface DMs were fabricated using this approach. Nevertheless, the tilted-front-interface approach will be developed further as soon as technology provides a better process for applying a high-quality wedge on the top of a multilayer.
6. Complementary pairs of DMs – were first mentioned by V. Laude [39] and later by F. Kaertner et al. [6]. This approach uses two different mirrors with identical in amplitude and periodicity GDD oscillations shifted with respect to each other by a half period, so that the averaged GDD oscillations, being half sum of them, are very small. For a pair of DMs covering <1 octave, such a design can be realized relatively easy. For broader wavelength range, the design and fabrication of complementary DM pairs is very challenging; nevertheless, in Ref. [28], a DM pair with controlled dispersion over 1.5 optical octaves, resulting in sub-3-fs pulses, has been demonstrated theoretically and experimentally. The currently available pulse limit of 1.5 optical cycles [28] can be pushed to a sub-optical cycle pulse by using broader DMs based on the complementary approach. To reduce the GDD oscillations, one can use three, four, and even more

independent mirrors. The main obstacle in wide use of this approach is the necessity to perform extremely accurate deposition runs perfectly matching each other.

7. Time-domain DMs – for the design of time-domain DMs, a criterion targeted at reaching shortest possible pulses with maximum possible energy at the output of a compressor containing such mirrors is used. This optimization criterion includes two parameters allowing one to adjust the relative weights of the mentioned targets with a high flexibility [7, 40, 41]. This approach allows efficient compression of pulses down to sub-5-fs durations. The time-domain approach has greater flexibility in controlling the pulse duration, and the pulse energy concentration, compared to conventional approaches, which use phase, GD, or GDD as target for optimization. If the incoming pulse spectrum and phase modulation are unknown or is not stable, the time-domain approach cannot be used.
8. Double-angle DM – is based on the combined usage of identical DMs at two different angles of incidence [13]. Owing to the angular shift of spectral characteristics, GDD curve oscillations come into antiphase for two carefully selected angles of incidence, thus, reducing residual oscillations. ‘Double-angle’ DMs offer: i) Better manufacturing stability compared to the complementary-pair approach and ii) reduced manufacturing costs compared to the complementary-pair approach, which requires two perfectly matched coating runs. Only one coating run is sufficient for the manufacturing of double-angle DM. Of course, double-angle DM provides higher residual level of averaged GDD oscillation compared to the complementary pairs approach.

All of these approaches have been successfully applied to visible and near-IR ranges. Their strengths and weaknesses are discussed in [30, 33, 42] at greater lengths.

In all of the cases, efficient thin film design software should be used in order to obtain designs with the specified performance. As design problems with GD/GDD requirements is among the most complicated ones, it is very important to use an analytical approach for the computation of GD and GDD, as well as of other values required in the implementation of numerical algorithms (e.g., the gradient of the merit function for optimization algorithms, the perturbation function for the needle variation approach, etc.). A special selection of starting designs can be very helpful in the course of finding a solution. For example, mirrors with layer optical thicknesses changing with layer number can be quite useful as a starting point when searching for a design of a dispersive mirror. Needle optimization [43] and gradual evolution [44] techniques allow to obtain a set of the best possible designs. Successive study of these multiple solutions [45] allows one to select a design, which is better suited for manufacture.

Below, we will present in detail two extreme cases located in short- and long-wavelength ranges.

4 UV mirror

UV ultrafast sources promptly become more and more important in the fields of biological and chemical research [46]. Introduction and technological verification of novel coating materials suitable for UV, such as HfO_2, allowed expanding the application of DMs to shorter wavelengths [27]. Significant advance has been made in generation of several tens of femtosecond pulses with a central wavelength below 350 nm using DMs [47]. Shortening further the pulse duration requires both further expansion of the spectra and possibilities of dispersion control over the wide bandwidths.

Necessity to work in the near-UV significantly decreases the range of materials, which can be used for dispersive coating production. For instance, the convenient and widely used because of their high refractive indexes TiO_2 (n_H~2.45) and Nb_2O_5 (n_H~2.35) cannot be implied, as both materials have pronounced absorption in the near-UV region. It has been shown that HfO_2 (n_H~2.05) is an appropriate material for dispersive coatings in the UV region [17]. Thus, HfO_2 was used as a high-index material, while keeping SiO_2 as a low-index material. However, achieving high reflectivity over large bandwidths with controlled dispersion properties using HfO_2/SiO_2 pair is challenging. The UV mirrors were produced with a magnetron-sputtering Helios machine (Leybold Optics GmbH, Alzenau, Germany). Helios is equipped with two proprietary TwinMags magnetrons and a plasma source for plasma/ion-assisted reactive middle-frequency dual-magnetron sputtering. The magnetrons are optimized for high sputtering rates and high optical layer performance. The system was pumped by turbomolecular pumps to 1×10^{-6} mbar before deposition. Argon and oxygen were used for both magnetrons. The purity of the Si target was 99.999% and that of the Hf target, 99.9%. By changing the electric power applied to the cathode, it was possible to vary the deposition rate. The film quality was found to degrade at too high deposition rates. A good layer quality was obtained at a rate of around 0.3 nm/s for HfO_2 and

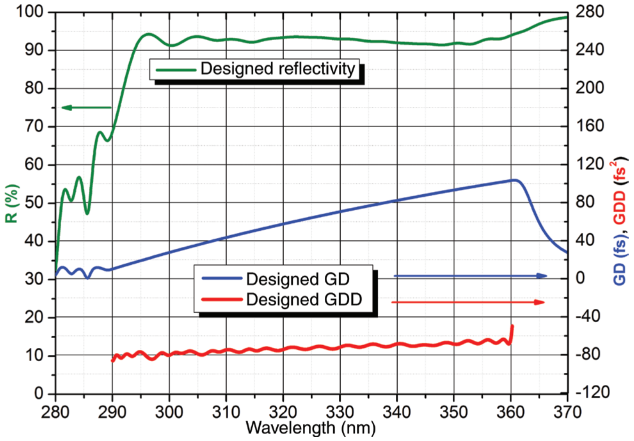

Figure 2 Designed reflectivity (green curve), GD (blue curve), and target GDD (red curve) at 45° AOI of the DM for 290 nm to 360 nm UV region.

at 0.4 nm/s for SiO_2. For the wavelength range below 380 nm, dielectric layers of HfO_2 deposited with magnetron sputtering technique provide weak absorption. The value of the extinction coefficient of HfO_2 at 300 nm is obtained as low as 2×10^{-4} by measuring the reflectivity and transmission of the multilayer stack and with a consequent reconstruction in OptiRE from OptiLayer software family [48]. For the SiO_2 layer, this value is 5×10^{-5}. As absorption scales with the thickness of the layer and the frequency of the incident light, decreasing the overall reflectivity of the multilayer stack, it is beneficial to design as thin a structure as possible. At the same time, in order to introduce significant GD in a broad spectral range, it is obligatory for DM to be thick enough in order to introduce sufficiently strong interference effects, which in turn sets the value of introduced absorption and limits potential reflectivity. In order to fulfill both requirements, an alternative path to the design of the multilayer stack has been taken. Namely, the presence of SiO_2 has been increased in order to reach the necessary thickness, while the presence of HfO_2 has been kept relatively low in order to suppress absorption.

A broadband dispersive multilayer mirror with high reflectivity in the near-UV region from 290 nm to 360 nm and average GDD of ~-75 fs^2 for a 45° angle of incidence (AOI) was recently realized [49]. The designed reflectivity, GD, and GDD curves are presented in Figure 2.

The created design consists of 86 consecutive layers of materials, reaches overall total physical thickness of 8.14 µm, and exhibits spectral averaged reflectivity of 92% in the target spectral range.

The mirror has been coated on fused silica substrate by means of magnetron sputtering technique, allowing us to reach a 0.5-nm accuracy in layer thicknesses. The measured GD and GDD data are presented in Figure 3 together with the corresponding designed data. The comparison of designed and measured GD represents evident correspondence in the whole working wavelength region. Successful dispersion management in the near-UV region achievable with this mirror paves the way to the generation of few femtosecond pulses with a central wavelength below 360 nm.

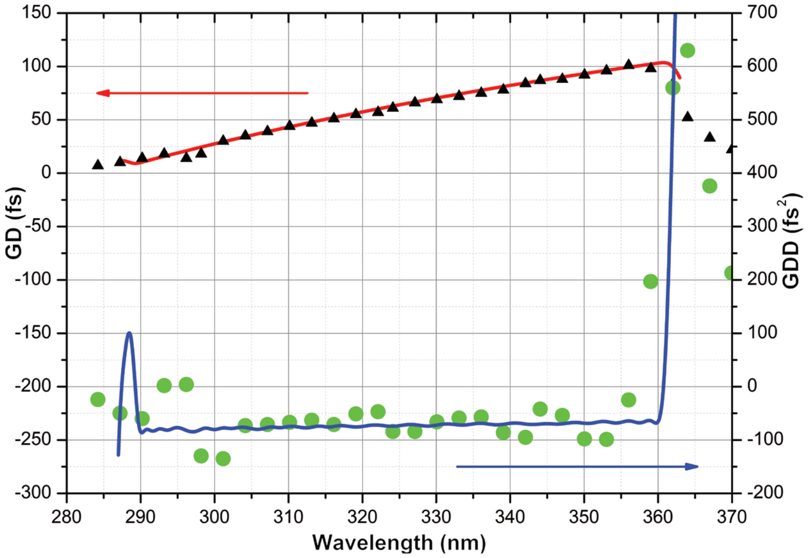

Figure 3 Designed GD at 45° AOI for p-polarized light (red solid curve) and measured GD at 45° AOI p-polarized light (black triangles); designed GDD at 45° AOI p-polarized light (blue solid curve) and measured GDD at 45° AOI p-polarized light (green circles) of the manufactured UV mirror.

5 IR mirror

The generation of ultrafast pulses in the near and mid-IR attracts more and more attention in connection with the quickly developing attosecond science. The implementation of near to mid-IR ultrafast sources to supercontinuum generation in hollow core fibers and gas cells [50, 51] will allow one to move the electron cut off energies further into the spectrum, thus, unchaining possibilities for generation of unprecedently short attosecond pulses, as well as the conventional generation of single attosecond pulses. As a probe step, a DM for the optical parametric oscillator (OPO), which produces two thirds-octave-wide spectrum centered at the subharmonic (3120 nm) of the femtosecond pump laser [26] was recently developed.

In Figure 4, the reflectivity and the GD of the designed DM are shown. This DM is used also as an input coupler and provides high transmission at 1560 nm for the pump wavelength. The design consists of 50 layers of Nb_2O_5/SiO_2. The IR mirrors were also produced with the Helios machine. For these mirrors, we installed Nb and Si targets in the cathodes. The electric power of the Si cathode is 4500 W, and the power of the Nb cathode is 3500 W. The power applied to the Nb cathode was not constant because it operated in the oxygen control (or lambda control) mode, which guaranteed stable film properties. The gas pressure was 1×10^{-3} mbar during the sputtering process. Oxygen was fed near the targets to oxidize the sputtering films. The distance from the targets to the substrates was 100 mm. The purity of the Si target was 99.999% and that of the Nb target, 99.9%. Deposition rates of around 0.4 nm/s were selected for both materials. The total optical thickness of the design reaches 41 μm.

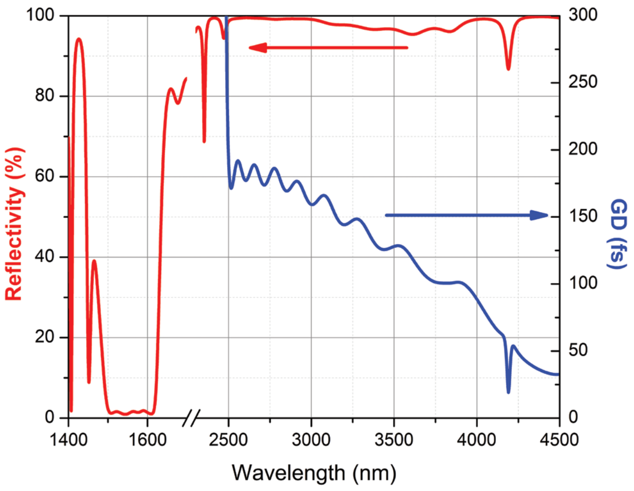

Figure 4 Reflectivity (red curve) and GD (blue curve) of the designed DM are shown.

Similar as in the case of UV mirrors, the IR DMs were produced using magnetron-sputtering technology. The produced mirrors have been measured using a resonance scanning interferometer [52]. The result from the GD measurement is shown in Figure 5. A convincing correspondence of measured and design curves in the range of 3000–4200 nm has been reported.

These mirrors have made it possible to demonstrate that the two OPOs show stable spatial and temporal interference in the wavelength region of 2500–4000 nm and are mutually locked in frequency and in phase [26].

6 Conclusions

For successful DM production, the layer thickness of the DM design has to be controlled with sub-nm accuracy during the deposition. This is especially true for UV and deep-UV spectral ranges, where it is also important to minimize possible interdiffusion of layer materials and roughness of layer boundaries. Additionally, refractive indices have to be kept homogeneous and stable for every layer of the design. The latter requirement is more difficult in the far-IR range, where the thickness of each layer and the total thickness of the design are necessarily large. As a result, the deposition time of thick coatings is large, and fluctuations of deposition parameters are more likely. Another problem, significant for thick IR coatings, is the

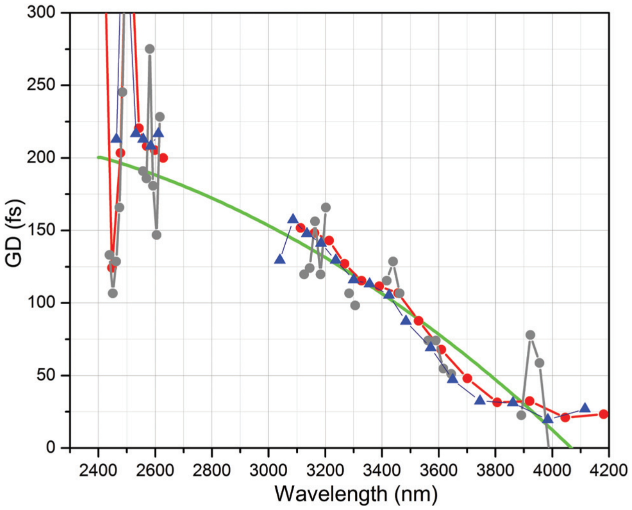

Figure 5 GD measurement of DM in the wavelength range of 2500–4200 nm. The green curve is the target GD adjusted to the delays in the measurement system. The red, gray, and blue curves are three independent GD measurements.

high level of stress, which decreases the stability and the performance.

We surveyed a set of dispersive mirrors, which are able to provide control of reflectivity and phase in a broadband range starting from 220 nm to 4500 nm. The working wavelength range is limited mostly by the properties of available layer-forming materials, namely, by absorptance of layers in the UV and far-IR ranges. The limit can be extended in the UV range down to 190 nm by using Al_2O_3 instead of HfO_2. The IR long-wavelength limit can be moved to 10 μm by using typical IR materials such as Si, Ge, ZnSe, and ZnS.

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