

# Recent development and new ideas in the field of dispersive multilayer optics

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A dispersive-mirror-based laser permits a dramatic simplification of high-power femtosecond and attosecond systems and affords promise for their further development toward shorter pulse durations, higher peak powers, and higher average powers with user-friendly systems. The result of the continuous development of dispersive mirrors permits pulse compression down to almost single cycle pulses of 3 fs duration. These design approaches together with the existing modern deposition technology pave the way for the manufacture of dielectric multilayer coatings able to compress pulses of tens of picoseconds duration down to a few femtoseconds. © 2010 Optical Society of America

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## 1. Introduction

The goal of generating short laser pulses down to the limit set by a single wave cycle of light has been pursued ever since the invention of lasers. Laser pulses consisting of only a small number of wave cycles allow more efficient exploitation of nonlinear optical effects [1] with implications as striking as the generation of single subfemtosecond light pulses [2]. The controlled superposition of light frequencies extending over more than one octave together with carrier-envelope phase control [3–5] pave the way for shaping the sub-cycle evolution of light fields in laser pulses. Here, we provide an overview of dispersive multilayer optics [6] offering high reflectivity and controlled group-delay dispersion over some 1.5 octaves spanning ultraviolet to near-infrared frequencies. Currently, we cannot imagine ultrashort pulses being obtained without dispersive multilayer optics. The dispersive mirror (DM) [6–25] is one of the key elements of most ultrafast (femtosecond) lasers. While being able to provide control of group-delay dispersion (GDD) over unprecedented bandwidths, broadband DM technology

suffers from unavoidable spectral oscillations of the GDD [26]. These oscillations may adversely affect the quality of the femtosecond laser pulses being controlled with the DMs.

The manufacture of DMs can be as challenging as their design. A DM is extremely sensitive to a discrepancy in layer thickness. Magnetron sputtering and ion sputtering technologies provide sufficient layer control precision in most cases. Modern sputtering technology can provide subnanometer precision in controlling the layer thickness. Some applications, such as highly DMs [21,25], require angstrom precision. The extreme sensitivity of the DM can then be reduced by applying a special, robust design algorithm [27].

The result of the 15 yr evolution of the design and fabrication of dispersive multilayers [7–25] allows the development of structures with low loss and high dispersion over a wide spectral range and permits compression down to the theoretical limit of the pulse.

## 2. Dispersive Multilayer Mirror

The main problem of DM design concerns unavoidable phase oscillations, which appear due to interface mismatch between the top layer of the multilayer structure and the external medium. The GDD oscillations lead to the destruction of the pulse shape.

Several approaches have been devised for suppressing these undesirable oscillations: double-chirped mirrors [7,9], back-side-coated DMs [12], complementary pairs of DMs [8,13,14,19,20], tilted-front-interface DMs [15], “Brewster angle” DMs [16,18], and double-angled DMs [24]. Recently, a time-domain approach based on the direct optimization of the parameters of the compressed optical pulse was also reported [17,22,23]. Chirped mirrors (CMs) rely on a multilayer structure with a gradual change in the optical thickness across the structure, resulting in a wavelength-dependent penetration depth of the incident radiation [6]. A CM is characterized by a certain value of the GDD, the second derivative of the phase shift on reflection with respect to the angular frequency [see Fig. 1(a)]. Alternatively, group delay (GD) variation may be introduced with resonant structures [or Gires–Tournois interferometer (GTI)] employing a wavelength-dependent storage time of the incident radiation [10,11] [see Fig. 1(b)]. Both effects may coexist [21,25] to improve the performance. In this case, multilayer mirrors should be called DMs.

A DM is a dispersive optical interference coating usually designed by optimizing the initial multilayer design. Optimization software (such as Optilayer [28], TFCalc [29], and Essential Macleod [30]) can be used to design the DM. During design optimization, residual GDD oscillations drop to a low level. The amplitude of the GDD oscillations determines the amount of energy that transfers into the satellite(s). The period of the oscillations in the spectral domain determines the position of the satellite in the temporal domain.

#### A. Resonance Structure or GTI Mirror

Two interfaces separated by an optical distance corresponding to the half-wavelength of the incident radiation resonantly enclose the impinging wave. Such nanoscale Fabry–Perot interferometers embedded in the multilayer structure can introduce large group delays at selected wavelengths, as shown in Fig. 1(b). The GTI approach is realized by optimizing the last few layers in the symmetrical quarter-wave layer stack (or Bragg structure). A standard quarter-wave stack or Bragg structure has zero GDD at the center of its stop band, with little variation over the entire high-reflectivity range. This results in a design exhibiting greater tolerance for manufacturing errors. The lower number of aperiodic layers means this approach leads to a structure with smooth GDD that exhibits little sensitivity to deposition errors.

For a broader bandwidth, multi-GTI can be used [10,11]. The only difference between a multiresonance and a single-resonance effect is that a number of cavities are organized in one multilayer structure. The multi-GTI mirror is characterized by a larger bandwidth and a large GDD value, compared to a single GTI mirror.

#### B. Double-Chirped Mirrors

The multilayer Bragg structure can be decomposed into a series of symmetric index steps, as is shown in Fig. 2. The depicted structure with an antireflection (AR) coating on the top can provide a smooth GDD characteristic, allowing compression of the pulse down to a duration of 6.5 fs [9]. AR makes it possible to match an interface mismatch between

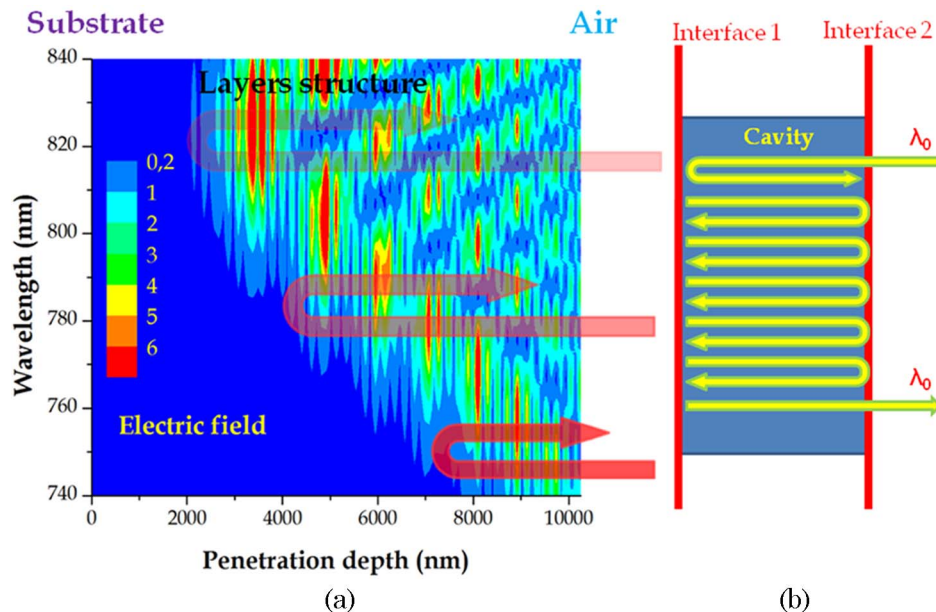


Fig. 1. (Color online) (a) Schematic explanation of the penetration effect. The optical thickness between different coating layers in the dielectric chirped mirror structure changes gradually. This causes different wavelengths to penetrate to a different extent and, hence, exhibit a GD. In this example, the shorter wavelengths (e.g., blue) are reflected near the surface of the multilayer mirror, while the longer wavelengths (e.g., red) penetrate deeper. (b) Schematic explanation of the resonance effect: When the optical thickness of the layers is close to half of the wavelength, they act as a cavity of a Fabry–Pérot interferometer.

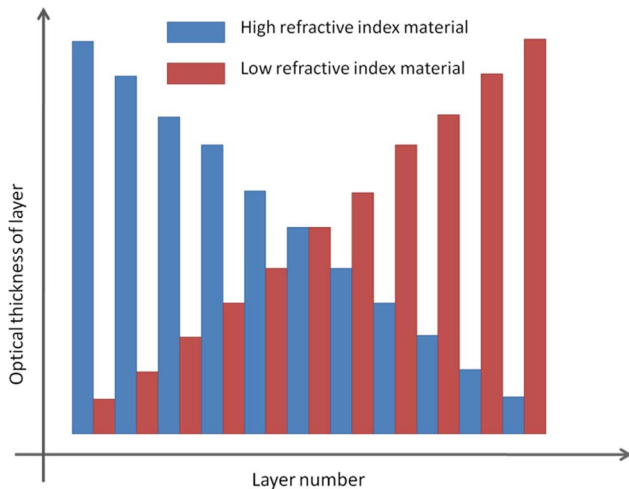


Fig. 2. (Color online) Layer structure of the double CM is depicted.

the top layer and the external medium, resulting in oscillation-free GDD characteristics.

Double CM has its limitations, as does any approach. The main limitation is a relatively narrow operating bandwidth. To overcome this bandwidth limitation and keep the GDD oscillations small, it is necessary to design broadband AR with a reflection below 0.1% over the entire bandwidth. The amount of light reflected by the AR coating mainly depends on the bandwidth and the maximal contrast of available coating materials. The number of materials is limited. The largest contrast in refractive indices can be obtained with  $\text{TiO}_2$  and  $\text{SiO}_2$  (2.4 and 1.45 at 800 nm, respectively). The maximal bandwidth of AR with a reflectivity  $<0.1\%$  [31–33] can be estimated to be  $\sim 300$  nm. As a result, one individual double CM cannot be used for compressing to sub-5 fs pulses, whose full bandwidth is  $>300$  nm.

### C. Brewster Angle CM

The elegant way of avoiding the cause of oscillations—interface mismatch (interaction of Fresnel reflection from the top layer and light penetrating into the multilayer stack)—is to use  $p$ -polarized light at the Brewster angle [15]. Placing a CM at the Brewster angle for the top layer ensures only a small amount of Fresnel-reflected light, resulting in oscillation-free GDD. Baum *et al.* [18] report the compression of a broadband spectrum corresponding to a pulse duration of a few cycles.  $P$ -polarized light at large angles (the Brewster angle of fused silica is  $\sim 56^\circ$ , for example) requires a significantly higher number of layers and greater total optical thickness. As a result, the manufacture of Brewster angle mirrors with a reflectivity  $>99\%$  becomes a problem. The intracavity beam bounces at a small incidence angle, which makes it unrealistic to use the Brewster angle CM as an oscillator mirror.

### D. Back-Side-Coated CM

In 2000, the back-side-coated CM was proposed by Matuschek *et al.* [12]. The authors propose using a

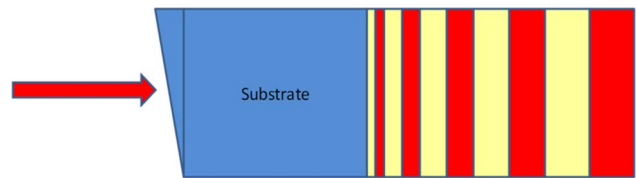


Fig. 3. (Color online) Sketch of the main principle of back-side-coated CM.

wedged substrate on one side. The irradiation penetrates through the wedged side of the substrate and interacts with a multilayer structure, as shown in Fig. 3. The wedge spatially splits the Fresnel reflection and the main part of the irradiation reflected from the layers. Such a design can be used to obtain a smooth GDD characteristic.

Unfortunately, the substrate must be a few millimeters thick to stay flat with a relatively thick dielectric multilayer on the top. The delay caused by reflection from the multilayer structure is neglected due to the delay caused by the beam passing through the substrate twice. Back-side-coated CM has smooth GDD, but it cannot be used for compression of a pulse due to the large path that the beam travels inside the substrate.

### E. Tilted-Front-Interface CM

It seems that the back-side-coated CM paved the way for the tilted-front-interface CM [15]. To avoid Fresnel reflection for the entire interface, we now attach the wedge on the top layer (see Fig. 4). The tilted-front-interface CM requires an additional AR coating on top of the wedge to minimize losses. The requirements for AR of the coating on the top of the tilted-front-interface CM are not as strict as for double CM. The amount of Fresnel-reflected light brings only additional losses, but it has no influence on the GDD oscillation.

The weakest point in the tilted-front-interface CM is the technology for applying a wedge onto the top of the multilayer stack. As far as we know, a tilted-front-interface CM with only a small diameter (half an inch) was fabricated using the tilted-front-interface approach. Nevertheless, in our opinion, the tilted-front-interface approach has potential as soon as the technology provides a process for applying a high-quality wedge on the top of the multilayer.

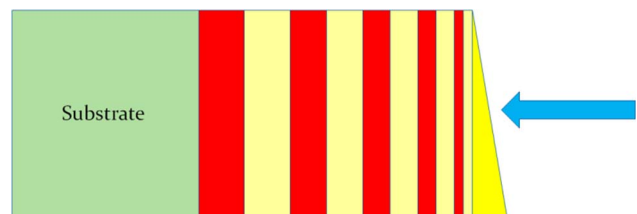


Fig. 4. (Color online) Tilted-front-interface CM. Light penetrates through the wedge on the top of the multilayer stack. The beams reflected by the multilayer and the wedge are spatially separate.

## F. Complementary Pairs of CMs

A first complementary pair was mentioned in Ref. [8], and later, Kaertner *et al.* [13,14] used two different mirrors for which the GDD curves have the same oscillations and are shifted by only half a period so that the average GDD curve has oscillations which are as low as possible (see Fig. 5). For a pair of CMs covering less than 1 octave, such a design can be realized easily.

In Ref. [20], the authors numerically and experimentally demonstrate a chirped mirror pair with controlled dispersion over 1.5 optical octaves. The complementary pair of such mirrors has average reflectivity >95% in a range between 400 and 1200 nm with a residual GDD oscillation <100 fs<sup>2</sup>, as shown in Fig. 5. The mirror pair makes it possible to compensate a chirp of the corresponding spectrum (with a smooth phase), resulting in sub-3 fs pulses.

In order to design a complementary CM pair, the following merit function can be used [19,20]:

$$F = \frac{1}{L} \sum_{j=1}^L \left( \frac{R_p(\lambda_j) - R^{(j)}}{\Delta R^{(j)}} \right)^2 + \left( \frac{\text{GDD}_p(\lambda_j) - \text{GDD}^{(j)}}{\Delta \text{GDD}^{(j)}} \right)^2, \quad (1)$$

where  $R_p(\lambda_j)$  and  $\text{GDD}_p(\lambda_j)$  are theoretical characteristics at wavelength  $\lambda_j$ ,  $R^{(j)}$  and  $\text{GDD}^{(j)}$  are target values,  $\Delta R^{(j)}$  and  $\Delta \text{GDD}^{(j)}$  are corresponding tolerances, and  $L$  is the number of selected wavelength points. The currently available pulse limit of 1.5 optical cycles [34] can be pushed to a suboptical cycle pulse by using broader DMs based on the complementary approach. To reduce GDD oscillations, one can use three, four, and even more independent mirrors.

## G. Time-Domain DM

A transform-limited short pulse, which can be calculated by the inverse Fourier transform of an

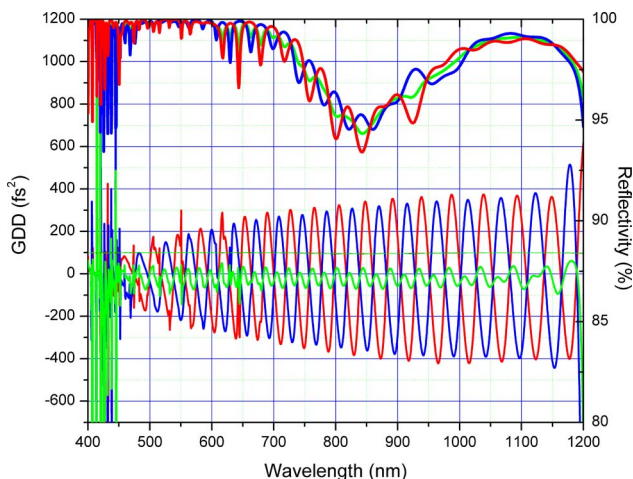


Fig. 5. (Color online) Calculated reflectivity and GDD of a pair of ultrabroadband CMs. The red curves represent one single mirror, the blue curves the other single mirror of the pair. The green curve is the average of the red and blue curves per bounce.

arbitrary input spectrum, propagates in a dispersive medium or a set of dispersive materials. The Fourier transform-limited pulse (FTLP) would be the shortest pulse that could be obtained with any design approach. The pulse stretches in dispersive media due to the dispersion of elements inside or outside the laser oscillator. A DM compressor can be employed to compensate this dispersion and thus compress the pulse. In the conventional design approach, the mirror dispersion must be as close as possible to the dispersion of the laser elements, but of the opposite sign. In the new time-domain design approach, the algorithm needs structures that allow the best possible comparability of the pulse after passage through the dispersive medium and the DMs (in Fig. 6).

In Refs. [17,22,23], authors report the results of the realization, application, and comparison of the two design approaches for sub-5 fs pulse compression. By changing the optimization parameters [22], the pulse duration can be traded off against the energy concentration. The time-domain approach allows us to control the pulse duration directly. There are two experimental requirements: (i) optimize the design so that the shortest pulse is generated, and (ii) optimize the design so that the maximum energy is concentrated in the main pulse (in this case, the pulse duration can be 10% longer). The time-domain approach has greater flexibility in controlling the pulse duration, and the pulse energy concentration has already been mentioned a couple of times.

## H. Double-Angle DM

In Ref. [24], the authors proposed a novel concept for suppressing GDD oscillations based on identical DMs used at two different angles of incidence in combination (Fig. 7). The “double-angle” DMs offer (i) better manufacturing stability compared to the conventional complementary-pair approach [8,13,14,19,20], and (ii) reduced manufacturing costs compared to the complementary-pair approach, which requires two perfectly matched coating runs. By properly selecting incidence angles and the application of the specialized version of the needle-optimization technique [35,36], we designed, manufactured, and characterized a double-angle DM with very low overall GDD oscillations. To demonstrate the utility of the double-angle approach, the mirrors were used for compressing pulses that had been spectrally broadened in a hollow fiber to a duration of 4.3 fs, which is very close to their Fourier limit of 4.2 fs (see Fig. 7).

## 3. Pulse Compression with the DM

Ti:sapphire femtosecond chirped-pulse amplification (CPA) lasers are in use all over the world. Their

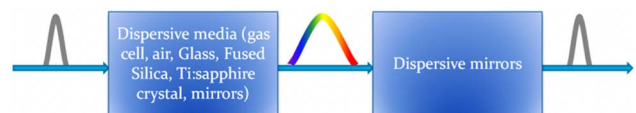


Fig. 6. (Color online) Basic scheme of time-domain target approach.

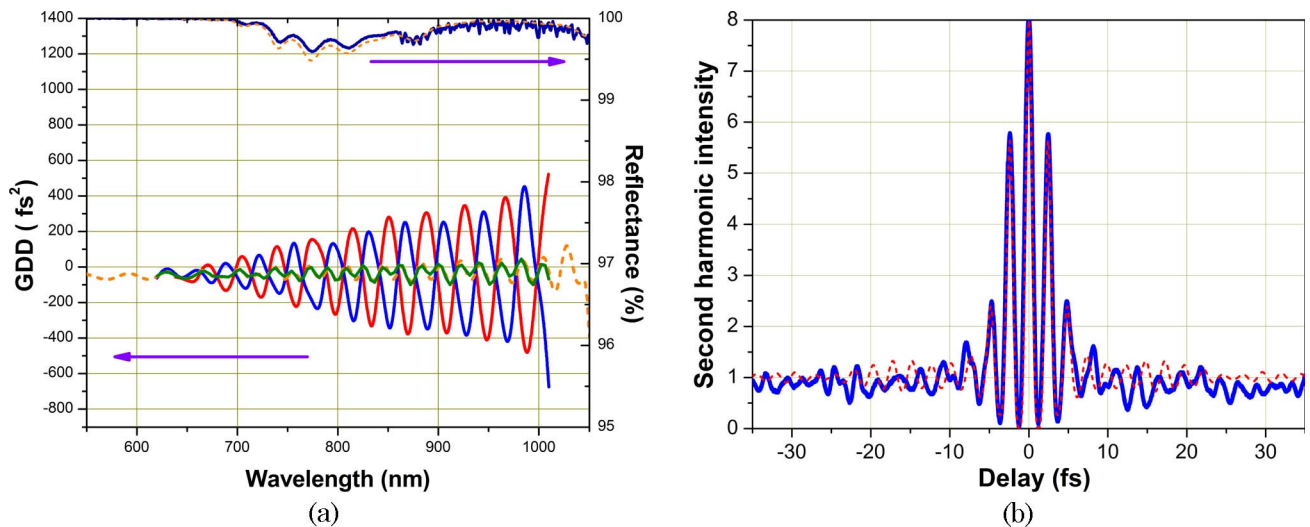


Fig. 7. (Color online) (Left) Measured GDD and reflectance of our prototypical double-angle DM for angles of incidence of 5 and 20° (solid lines). GDD and reflectance of the theoretical design are shown by dashed lines. Red and blue curves correspond to angles of incidence of 5 and 20°, respectively. Dark green and dark blue curves show the average measured GDD and reflectance. (Right) Dashed line: Theoretical interferometric autocorrelation of a pulse with the frequency spectrum is shown in the absence of spectral phase modulation. This Fourier-limited pulse has a duration (FWHM) of 4.2 fs. Solid line: Measured interferometric autocorrelation function of the pulses compressed with our prototypical double-angle DM compressor, indicating a pulse duration of  $\sim 4.3$  fs.

operation relies on stretching and recompressing the pulses before and after amplification. Currently, complex, rather lossy, and alignment-sensitive optical systems based on prisms and/or diffraction gratings are employed for this purpose. Authors [21,25] demonstrate that the required dispersion of the order of  $10^4$ – $10^5$  fs<sup>2</sup> can be introduced by a set of high-dispersive chirped multilayer dielectric mirrors offering several advantages. Recent progress in the design and manufacture of multilayer high-dispersive mirrors (HDMs) has resulted in a quantum leap in their performance (reflectance and amount of the GDD), and it paves the way for a dispersive-mirror compressor for CPA. HDM exploits penetration and resonance effects simultaneously, thereby combining the advantages of chirped mirrors [6] and GTI structures [10,11], respectively.

The prototypical dispersive-mirror compressor has been designed [21,25] for a kHz Ti:sapphire amplifier and, in a proof-of-concept study, it yielded millijoule energy, sub-20 fs, 790 nm laser pulses with an overall throughput of  $\sim 90\%$  and unprecedented spatio-temporal quality. Dispersive-mirror-based CPA permits a dramatic simplification of high-power lasers and affords promise for their further development to shorter pulse durations, higher peak powers, and higher average powers with user-friendly systems. CPA implemented with DMs is therefore intrinsically free from angular chirp, pulse-front tilt, and nonlinear effects, with the added benefit of higher-order dispersion control. These novel, low-loss, and HDMs provide per reflection a GDD of  $-500$  fs<sup>2</sup> with  $<0.2\%$  loss for a wavelength range of 735–845 nm (see Fig. 8). Residual GDD oscillations in an HDM compressor imprint a (peak-to-peak) spectral phase oscillation of  $\sim 1.4$  rad on the compressed pulses.

These oscillations can be largely removed by the DAZZLER (Acousto-Optic Programmable Dispersive Filter) without compromising its throughput. We assessed our HDM design by means of standard computational error analysis. The layer thicknesses of a given design are randomly varied with 1 nm standard deviation. This value corresponds to the accuracy of our state-of-the-art magnetron sputtering machine (Helios, Leybold Optics GmbH, Alzenau, Germany).

They can be designed to provide GDD of either sign and, hence, compensate material dispersion both in the visible/near-infrared and the mid-infrared spectral range, where the dispersion of most materials changes its sign.

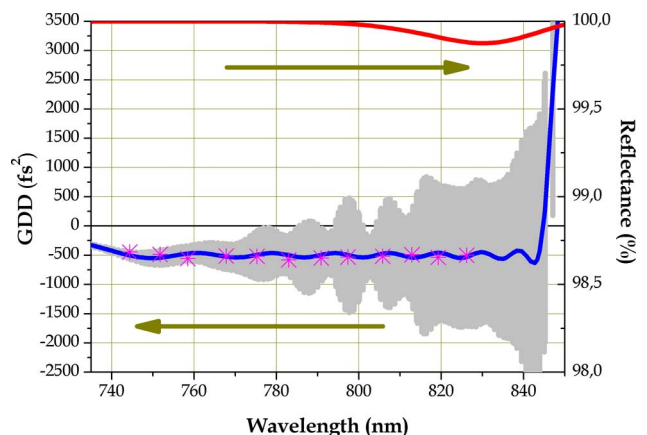


Fig. 8. (Color online) Calculated reflectance (red) and GDD (blue) curve of HDM for the design angle of incidence of 10°. Measured GDD with a white light interferometer (magenta) is in good agreement with calculated values. The gray area represents the probable range of GDD values.

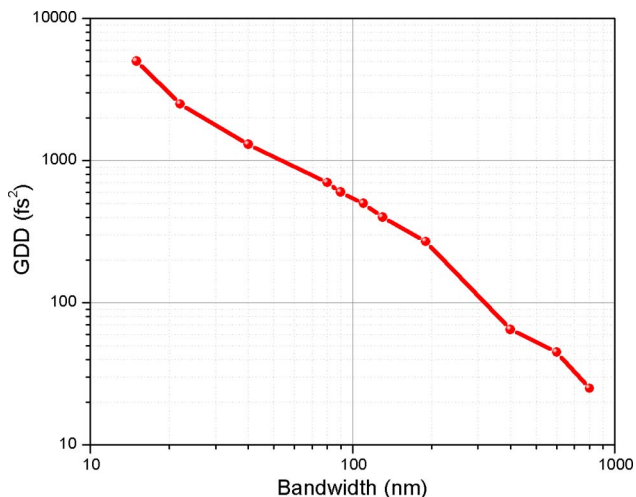


Fig. 9. (Color online) The highest absolute value of GDD as a function of the relative bandwidth, obtained at center design wavelength of 800 nm. The sign of GDD presented here is negative, but it can be positive, as well. The lines connecting the points serve as a guide to the eye.

#### 4. Highest Value of GDD

To summarize the results in Sections 2 and 3, we demonstrate that the broader the spectrum for which the GDD must be controlled, the lower the nominal value of the GDD that can be achieved. In other words, relaxing the requirement on bandwidths allows for higher values of the GDD. This finding has been inferred from the manufacture of 11 different DMs. In Fig. 9, each point represents the highest absolute value of the GDD obtained for a certain spectral width at the central wavelength of 800 nm. The actual values of the points along the GDD axis depend on the total optical thicknesses of the coating, layer materials, and required reflectivity. We try to present design for fair comparison with similar total optical thickness in the range from 14 to 18  $\mu\text{m}$ . The data provide a rule of thumb for the trade-off between the bandwidth and the dispersion achievable. Note that there is yet another trade-off to be appreciated, namely, the one between the highest achievable GDD and the reflectivity.

#### 5. Conclusion and Discussion

We believe that existing approaches are still not at their limit. The design approaches mentioned can be successfully used for a specific application. There is no universal approach. Another material combination with higher refractive index contrast and better accuracy of the deposition process together with further development of the mathematical design approaches could push the now-existing frontier toward shorter pulse durations, higher peak powers, and higher average powers. Another alternative for better performance may be a combination of different approaches: for example, the double-angle DM approach with time-domain optimization, or any other combinations. In the near future, dispersive multi-

layers will continue to play a critical role in the field of ultrafast physics.

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