

# Synthesis and manufacturing the mirrors for ultrafast optics

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## ABSTRACT

Properties of 3 types of multilayer dielectric chirped mirrors manufactured with Helios Leybold system (magnetron sputtering with plasma/ion assisted technology) are discussed. The first type includes mirrors providing negative group delay dispersion (GDD) and high reflectance in a wavelength range 650–1150 nm. Such mirrors are used in a broadband Ti:Sapphire oscillator for generating sub-6 femtosecond pulses. The second type of mirrors has extremely low both GDD oscillations and losses in a range 740-840 nm. Such mirrors are of interest for both long-cavity high-energy Ti:Sapphire oscillators and so-called external cavities for storing energy from femtosecond oscillators. The lowest residual GDD fluctuations achieved for this type of mirrors are 5 fs<sup>2</sup>. The third type is high reflectors with very high reflectivity. Different coating materials: Nb<sub>2</sub>O<sub>5</sub>, TiO<sub>2</sub>, Ta<sub>2</sub>O<sub>5</sub> together with SiO<sub>2</sub> are compared for these 2 types of mirrors. The results of reproducibility of such mirrors and their characterization in terms of reflectivity, losses, surface quality are presented. Comparison of the design with the manufactured mirror is also done.

**Key words** — coatings, mirrors, thin films, group delay dispersion, ultrafast optics.

## 1. INTRODUCTION

Dispersion-controlled or chirped mirrors (CM) were developed in 1994 <sup>1</sup> and offer broadband feedback and dispersion control in ultrafast Ti:sapphire systems by imparting negative dispersion over broad spectral range <sup>2,3</sup>. CM can compensate GDD of materials used in laser oscillators for generating femtosecond pulses at slightly negative dispersion or heavily chirped picosecond pulses at slightly positive net cavity dispersion. CM provides precise control of optical delay upon reflection together with high reflectivity over the desirable spectral range. Usually, CM has pronounced GDD oscillations for longer wavelength range. The oscillations occur due to an effective Gires-Tournois interferometer <sup>4</sup>. A double Gires-Tournois interferometer has been utilized to solve this problem <sup>5</sup>. It consists of symmetrical  $\lambda_0/4$  layers with only few last optimized layers. Such a design is not sensitive to small discrepancies in thickness of layers.

The complementary mirror approach is utilized in a case when the spectral range should be broad. The broadband dielectric multilayer mirror designs have been obtained by computer optimization and have a bandwidth of 500 nm at the central wavelength of 800 nm. One of the main problem with optimization of multilayer dielectric coatings is that the merit function usually has many local extrema, <sup>6</sup> so that there is always a risk that a local optimization routine will get stuck far away from the absolute extremum. In addition, GDD of a chirped mirror is highly sensitive to small discrepancies in a layer thickness of a calculated design and those of the manufactured mirrors. Global optimization was recognized a long time ago as a solution to these problems.

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We designed CM by using the memetic approach to global optimization that showed good results<sup>7</sup>. The stochastic quasi-gradient algorithm can be applied to reduce the mirror sensitivity to random perturbations of layer thicknesses that are introduced by any manufacturing process.

The aim of this paper is to present the dispersion and reflection characteristics of the chirped mirrors produced with the Helios sputtering machine. Performance of two laser oscillators equipped with the manufactured mirrors is also presented.

## 2. EXPERIMENTAL

The CMs were manufactured by Helios Leybold Optics sputtering system. The Helios is equipped with two of proprietary TwinMags magnetrons and a plasma source for plasma/ion assisted reactive middle frequency dual magnetron sputtering. The magnetrons have been optimised for high sputter rates and high optical layer performance. It takes only seven hours to produce one CM even for complex chirped multi-layer systems. Due to the high deposition rates  $\sim 0.4$  nm/s for Nb<sub>2</sub>O<sub>5</sub> and SiO<sub>2</sub>, the fast production is enabled. In case of ion beam sputtering the rate is lower, about 0.1 nm/s. The combination of Nb<sub>2</sub>O<sub>5</sub> and SiO<sub>2</sub> were chosen for broadband CMs as a compromise between the TiO<sub>2</sub> (high losses) and the Ta<sub>2</sub>O<sub>5</sub> (lower losses than for Nb<sub>2</sub>O<sub>5</sub> but also lower refractive index that leads to the narrower spectrum). In a case when relatively narrowband but low-loss mirror is necessary, Ta<sub>2</sub>O<sub>5</sub> can be used.

Important to note that the refractive indexes of Nb<sub>2</sub>O<sub>5</sub> ( $n=2.35$ ) and SiO<sub>2</sub> ( $n=1.48$ ) is higher than those achievable with electron beam evaporation, indicating very dense layers. 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.

In both cases structures of mirrors consist of two different materials with high (H) and low (L) refractive index. The actual refractive index of the layers must be determined just before the manufacturing process starts and then the existing design of a mirror has to be re-optimised with new refractive index values. To determine the refractive index and physical thickness of a single layer, its transmission spectral curve was measured and then the Optilayer software was used. The method of determining the optical properties of a thin film from a transmission measurement is not very precise, but fast enough. The accuracy of this method in determining the refractive index and physical thickness is  $\sim 0.5\%$  and sufficient for our application. Values of an actual refractive index and physical thickness of a film are necessary when a CM is in the manufacture process with the time control. In the case of CM, the optical control becomes difficult or even impossible.

The reflection spectra were measured with spectrophotometer PerkinElmer (Lambda 950). The reflection  $\leq 99.9\%$  was measured with the additional absolute reflection accessory (TNO). Reflection of higher values were measured with tunable Ti:sapphire laser by using ringdown technique. GDD was determined by using white light interferometer with the spectral accuracy 10 nm and dispersion accuracy  $\sim 10$  fs<sup>2</sup>. One-mirror measurement together with the data retrieval takes  $\sim 1$  h. A Fizeau interferometer was used for the surface quality measurements. The accuracy of the surface flatness determination is about  $\lambda/10$ . The properties of all multilayer dielectric structures are calculated by the commercial Optilayer software. For mirror designing, the Optilayer software together with the developed optimization algorithm<sup>7</sup> have been used. The advantage of the algorithm is that it allows designing a pair of complementary mirrors.

## 3. RESULTS AND DISCUSSION

### 3.1 The broadband chirped mirror

The dispersion characteristics of the input coupler mirror (IC3) and the complementary mirror (CM3) for broadband Ti:sapphire oscillators for producing 6 fs phase-stabilized pulses are shown in Fig.1.

The mirrors IC3 and CM3 have high reflectance (over 99%) in a wide range 620-1130 nm. Additionally, IC3 has high transmission (99 %) at the pump wavelength 532 nm. The IC3 and CM3 must be used together to make the resultant residual GDD oscillations shallow. It means that the GDD oscillations of each mirror must be controlled within 2 nm. The physical thicknesses of the IC3 and CM3 layers are shown in Table.1. The physical thicknesses of layers vary from 20 nm to 210 nm that makes it difficult to manufacture close to the design. The thickness of layers should be re-optimized always when the refractive index of materials changed. There are several technical sources leading to variations of the refractive index of the order of 1%.

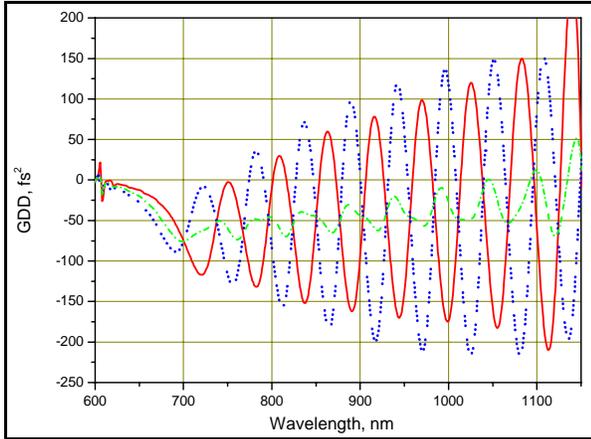


Fig. 1. Computed GDD as a function of the wavelength for the input coupler IC3 (solid line), the complementary CM3 (dashed line) and the averaged GDD of the input coupler IC3 and the complementary CM3 mirrors (dot line).

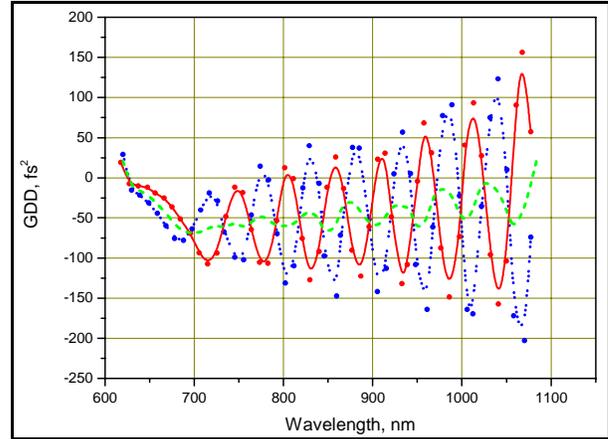


Fig. 2. Measured GDD as a function of the wavelength for the input coupler IC3 (solid line) and the complementary CM3 (dashed line) and averaged GDD of the input coupler IC3 and the complementary CM3 mirrors (dot line).

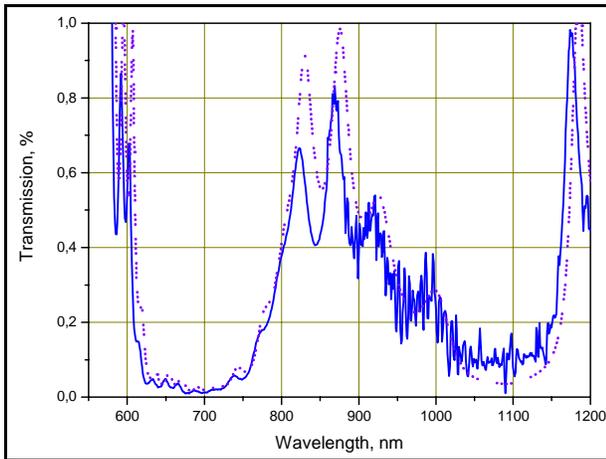


Fig. 3. Computed transmission curve of the CM3 mirror (dashed line) and the measured curve (solid line).

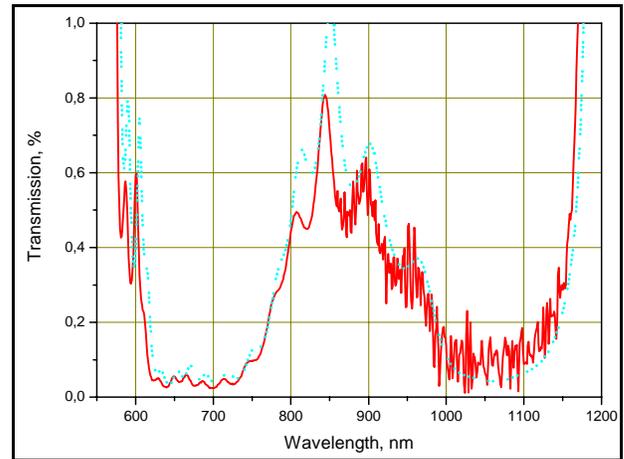


Fig. 4. Computed transmission curve of IC3 mirror (dashed line) and the measured curve (solid line).

Comparison of the GDD curves in Figs. 1,2 and the transmission curves in Figs. 3,4 shows that the manufactured mirrors have characteristics very similar to the design. The difference can be explained mostly by errors of the measurements. We have found that for quick testing a newly manufactured mirror one can compare the transmission curves of the design and the manufactured mirror. If they are identical, as in Figs. 3,4, the GDD is also close to the design. Indeed, one can see in Figs. 1 and 2 that the measured GDD curves are close to the computed ones. Nevertheless, a detailed inspection shows that the oscillation amplitudes of the measured curves in Figs.1, 2 are slightly lower than the computed ones. The measured reflection value  $R$  exactly follows the relation  $R=1-T$ , where  $T$  is the transmission, shown in Figs. 3,4. It means that the absorption is below our experimental detection accuracy. Therefore, the reflection curves are not presented.

Layer number	Material	The physical thickness of IC3 layer, nm	The physical thickness of CM3 layer, nm
1.	SiO <sub>2</sub>	197,61	197,84
2.	Nb <sub>2</sub> O <sub>5</sub>	154,7	149,94
3.	SiO <sub>2</sub>	208,78	208,43
4.	Nb <sub>2</sub> O <sub>5</sub>	139,48	132,38
5.	SiO <sub>2</sub>	215,74	207,51
6.	Nb <sub>2</sub> O <sub>5</sub>	133,05	130,43
7.	SiO <sub>2</sub>	201,84	192,64
8.	Nb <sub>2</sub> O <sub>5</sub>	126,66	124,33
9.	SiO <sub>2</sub>	208,15	194,85
10.	Nb <sub>2</sub> O <sub>5</sub>	134,37	128,05
11.	SiO <sub>2</sub>	191,1	189,76
12.	Nb <sub>2</sub> O <sub>5</sub>	123,93	124,13
13.	SiO <sub>2</sub>	191,63	181,8
14.	Nb <sub>2</sub> O <sub>5</sub>	121,94	119,08
15.	SiO <sub>2</sub>	185,11	179,95
16.	Nb <sub>2</sub> O <sub>5</sub>	121,86	117,92
17.	SiO <sub>2</sub>	180,77	176,74
18.	Nb <sub>2</sub> O <sub>5</sub>	117,22	113,58
19.	SiO <sub>2</sub>	175,79	171,68
20.	Nb <sub>2</sub> O <sub>5</sub>	113,32	110,78
21.	SiO <sub>2</sub>	171,71	168,51
22.	Nb <sub>2</sub> O <sub>5</sub>	110,9	108,97
23.	SiO <sub>2</sub>	168,79	166,1
24.	Nb <sub>2</sub> O <sub>5</sub>	109,22	107,44
25.	SiO <sub>2</sub>	166,4	163,53
26.	Nb <sub>2</sub> O <sub>5</sub>	107,66	105,24
27.	SiO <sub>2</sub>	163,94	159,13
28.	Nb <sub>2</sub> O <sub>5</sub>	105,58	100,18
29.	SiO <sub>2</sub>	159,67	145,6
30.	Nb <sub>2</sub> O <sub>5</sub>	100,99	86,66
31.	SiO <sub>2</sub>	147,85	130,73

32.	Nb <sub>2</sub> O <sub>5</sub>	89,03	90,44
33.	SiO <sub>2</sub>	130,28	144,08
34.	Nb <sub>2</sub> O <sub>5</sub>	88,03	93,52
35.	SiO <sub>2</sub>	141,65	137,86
36.	Nb <sub>2</sub> O <sub>5</sub>	93,1	81,86
37.	SiO <sub>2</sub>	138,16	117,74
38.	Nb <sub>2</sub> O <sub>5</sub>	84,47	82,04
39.	SiO <sub>2</sub>	121,47	134,14
40.	Nb <sub>2</sub> O <sub>5</sub>	81,35	87,89
41.	SiO <sub>2</sub>	129,91	121,99
42.	Nb <sub>2</sub> O <sub>5</sub>	85,5	71,65
43.	SiO <sub>2</sub>	124,66	114,4
44.	Nb <sub>2</sub> O <sub>5</sub>	77,43	82,68
45.	SiO <sub>2</sub>	115,91	127,25
46.	Nb <sub>2</sub> O <sub>5</sub>	78,82	75,96
47.	SiO <sub>2</sub>	120,8	98,34
48.	Nb <sub>2</sub> O <sub>5</sub>	78,2	72,88
49.	SiO <sub>2</sub>	113,1	124,54
50.	Nb <sub>2</sub> O <sub>5</sub>	73,36	80,72
51.	SiO <sub>2</sub>	110,49	92,33
52.	Nb <sub>2</sub> O <sub>5</sub>	75,39	63,09
53.	SiO <sub>2</sub>	114,05	121
54.	Nb <sub>2</sub> O <sub>5</sub>	72,38	83,97
55.	SiO <sub>2</sub>	98,46	83,54
56.	Nb <sub>2</sub> O <sub>5</sub>	70,34	55,01
57.	SiO <sub>2</sub>	116,07	126,26
58.	Nb <sub>2</sub> O <sub>5</sub>	73,75	88,86
59.	SiO <sub>2</sub>	78,59	49,08
60.	Nb <sub>2</sub> O <sub>5</sub>	69,14	65,7
61.	SiO <sub>2</sub>	131,63	155,07
62.	Nb <sub>2</sub> O <sub>5</sub>	54,41	43,61
63.	SiO <sub>2</sub>	53,16	47,77
64.	Nb <sub>2</sub> O <sub>5</sub>	110,82	109,8
65.	SiO <sub>2</sub>	161,37	153,52

Table 1. Layer thicknesses of the IC3 and CM3 designs.

### 3.2 Smooth-dispersion mirror

A CM for a high-energy long cavity Ti:sapphire oscillator (LCO) is designed to perform both the high reflection and the dispersion compensation of intracavity air, fused silica and Ti:sapphire in a range 740-840 nm. The distribution of refractive index of layers of LCO mirror is shown in Fig.5. LCO mirror consists of 17 pairs of alternating  $\lambda_0/4$   $\text{Nb}_2\text{O}_5$  and  $\text{SiO}_2$  layers and 5-chirped layers. The design can be presented as S (HL)<sup>17</sup>H 0.663496L 1.848203H 1.559812L 1.531112H 2.380152L A, where H denotes the high refractive index ( $n_H=2.35$  at 500nm) material  $\text{Nb}_2\text{O}_5$ , L is the low refractive index ( $n_L=1.48$  at 500nm) material  $\text{SiO}_2$ , S is the substrate (BK7) and A is air.

The spectral GDD and transmission of LCO are shown in Figs. 6, 7. As it can be seen, the designed GDD exhibits shallow oscillations, with deviations less than  $\pm 2 \text{ fs}^2$  from the required (approximately linear) function over the spectral range.

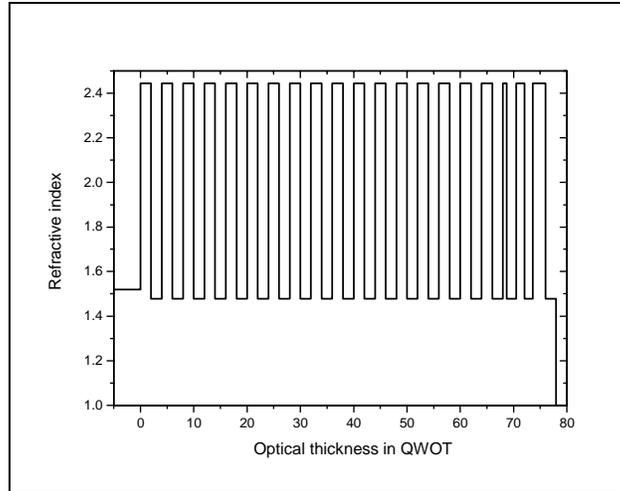


Fig. 5. Distribution of the refractive index of LCO mirror versus optical thickness of layers in the quarter wave length units.

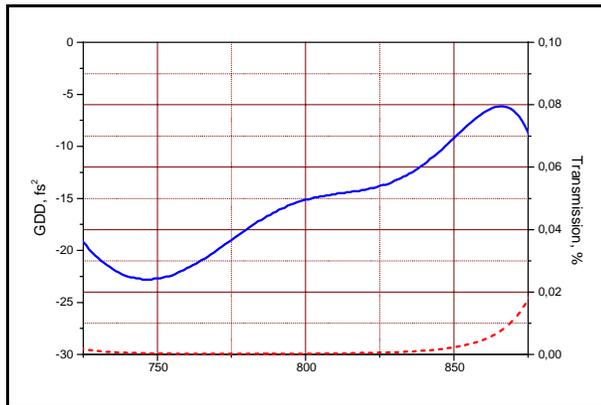


Fig. 6. Computed GDD (solid line) and transmission (dashed line) as a function of the wavelength of LCO mirror.

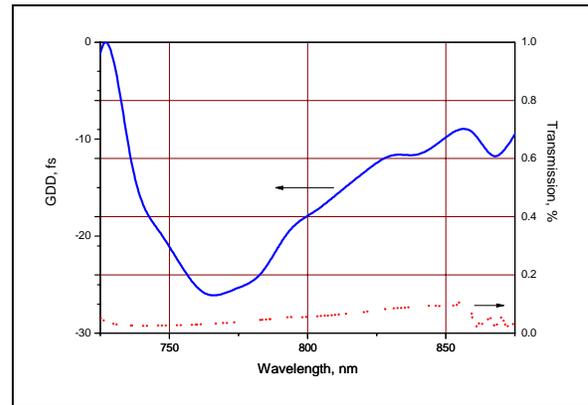


Fig. 7. Measured GDD and transmission versus wavelength of LCO mirror.

For the bandwidth  $\sim 100$  nm, we can avoid using the complementary mirror approach. The advantages of the proposed LCO mirror are high reflection and smooth GDD in the wavelength range 740-840 nm, see Fig. 6. As distinct in the design from a broadband CM, the LCO mirror can be based on a high reflector, with only few last layers optimized. The high reflector mirror usually consists of 10-20 pairs of alternating  $\lambda_0/4$  layers of high and low refractive index materials. A range where such a mirror has a stop-band, also corresponds to a smooth GDD around zero. The design is not as sensitive to errors as that for broadband chirp mirrors IC3 and CM3. The reflection of LCO is  $>99.9\%$  in a wavelength range 720-880 nm, as can be determined from Fig. 8. The average output power of a long-cavity Ti:sapphire

oscillator equipped with LCO mirrors increases ~10% in comparison to the oscillator with mirrors made with the “traditional” CM technique. LCO mirror was coated on a 2” substrate. The inspection has been done over all the surface of the mirror because of a special multi-pass geometry of a laser cavity (see inlet in Fig.12 below).

### 3.3 High reflector

The high reflector design which consists of alternating quarter wavelength, 40 layers of high refractive index material ( $\text{Nb}_2\text{O}_5$ ) and low refractive index ( $\text{SiO}_2$ ) has reflection as high as 99.99% in the wavelength range 720-880 nm. When the reflection of this LCO design is still not enough, instead alternating 50 quarter wavelength layers can be used with the last few layers optimized. The highest achievable reflectivity is determined by the absorption and scattering losses of the used materials. The comparison of the highest achievable reflectivity in a spectral range  $\geq 100$  nm with other coating techniques can not be easily done mostly because of lack of reliable information <sup>8</sup>.

### 3.4. Mirror reproducibility and the surface quality

Reproducibility of the manufacture process is demonstrated in Fig. 8 on the example of GDD. CM5 Mirror has the same design as CM6, but was manufactured in another day. The transmission curves have similar behavior including the fine features like shown in Figs.3 and 4. The spectral transmission reproducibility is within 5 nm. GDD is more sensitive to the errors, therefore it is shown in Fig.8 for the two mirrors. The observable shift in GDD curves can be explained by error in the layer thickness and uncontrollable changes in refractive index of materials during the production. Nevertheless, the reproducibility is good for such a complex design, at least we were unable to find comparable data in literature.

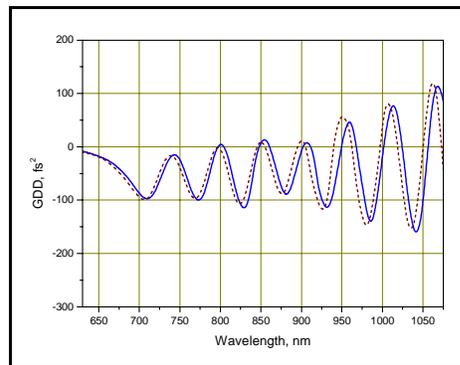


Fig. 8. Measured GDD as a function of the wavelength for CM5 mirror (solid line) and CM6 mirror (dashed line).

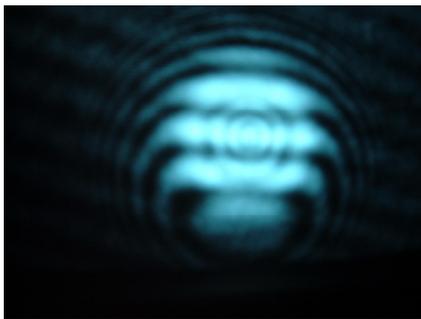


Fig. 9. The interferometer pattern of the BK7 substrate surface before coating.

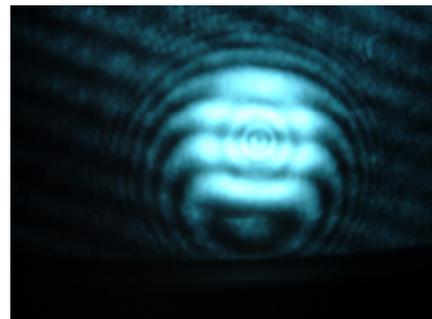


Fig. 10. The interferometer pattern of the coated BK7 substrate surface.

For all the mirrors, the substrates with the surface quality  $\lambda_0/10$  were used (Layertec, Optilab). The quality of uncoated and coated substrates was measured. In both cases the comparable results were obtained, i.e. the quality of a coated substrate (multilayer CM) can be also described as  $\lambda_0/10$ . The surface quality was determined from the

interferometer pattern shown in Fig.9 (uncoated substrate BK7) and Fig.10 (CM3 on BK7). The interferometer patterns for IC3, CM3, CM5, CM6 and LCO have equal quality. The diameter of our substrates was 25 mm and the thickness 6.35 mm. It means that the ratio thickness/diameter is 1:4. If the ratio is less (an example: 50-mm-diameter substrate of 9.5 mm thickness), the surface quality degrades after coating. Fused silica substrates demonstrate similar results.

We also checked the adhesion and the surface quality of multilayer coatings on GaAs wafers and found that it is possible to produce high-quality structures on such substrates. Coated-GaAs-related structures are useful as saturable absorber mirrors (SBR) in Ti:sapphire femtosecond oscillators (see Fig.12 below). Such special mirrors help starting the mode-locking regime and keeping it stable. Coating on GaAs structure allows to get the bandwidth of such a mirror around 100 nm.

## 4. LASER PERFORMANCE

### 4.1 Broadband femtosecond oscillator

Broadband mirrors IC3, CM3, CM5, CM6 were implemented into a broadband Ti:sapphire oscillator. Fig.11 shows the oscillator layout and Fig.12 - the generated spectrum.

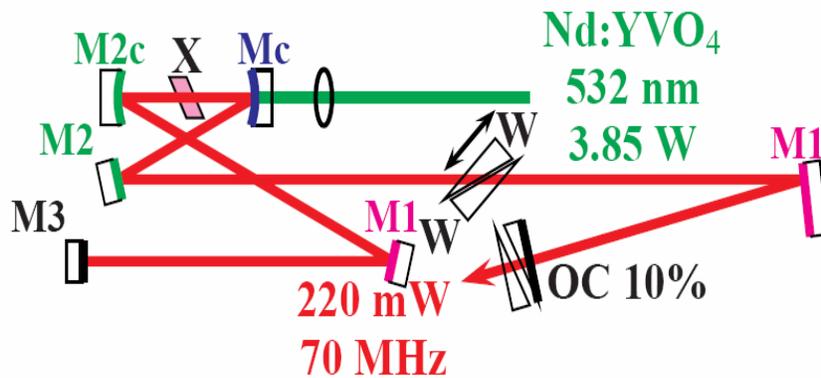


Fig.11. Layout of a broadband Ti:sapphire oscillator <sup>9</sup>.

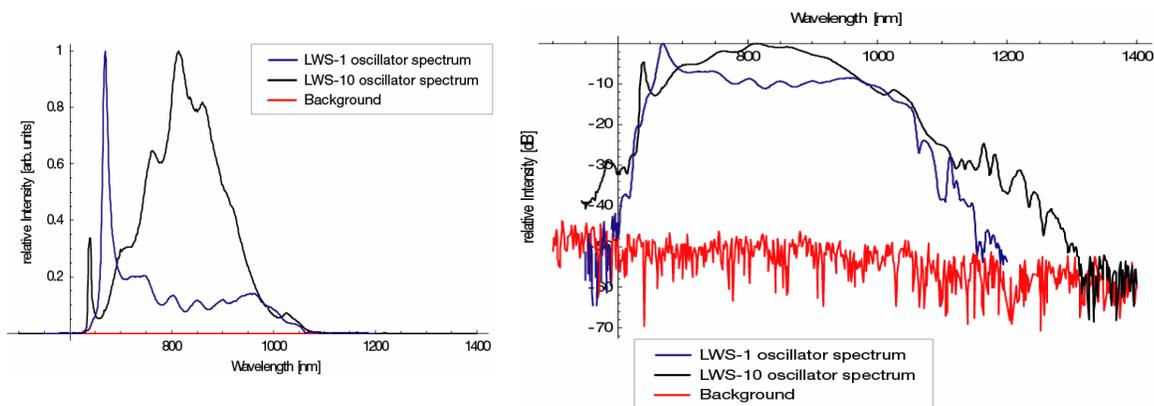


Fig.12. The generated spectrum out of the oscillator in linear scale (left) and logarithmic (right).

The difference of the realized oscillator with the known broadband Ti:sapphire oscillators demonstrated so far<sup>10,11</sup> consists in generation of compressible spectrum with high-energy components at the infrared part for a new type of a phase stabilization scheme<sup>9</sup>.

### 4.2. High-energy long-cavity oscillator

LCO mirror was manufactured for a high-energy long-cavity Ti:sapphire oscillator and for the enhancement cavity (to store the pulse energy from the oscillator). In the former case, the oscillator operates at so-called positive net dispersion regime<sup>12,13</sup>. The regime is extremely critical to the spectral shape of the dispersion. “Wrong” spectral dispersion leads to narrow spectrum and prevents laser operation at high pulse energy level. “Right” smooth GDD design allows generating spectrum up to 100 nm at 10 MHz repetition rate at 200 nJ pulse energy<sup>12</sup>. Our recent result is presented in Fig.13, showing the layout of 500 nJ, 2-MHz Ti:sapphire oscillator together with the net-cavity GDD and the generated spectrum. Worthy noting that it is more difficult to provide smooth dispersion for a longer oscillator cavity (of a fixed footprint). The reason for that is that for the laser compactness, one has to use more delay lines (two in case of 500 nJ oscillator in Fig.13) with the increased number of bounces on the mirrors. It means, that even weak deviations of GDD from the target value, being multiplied many times ( $12 \times 2 \times 2 = 48$  in our case), will result in big chaotic jumps of the dispersion.

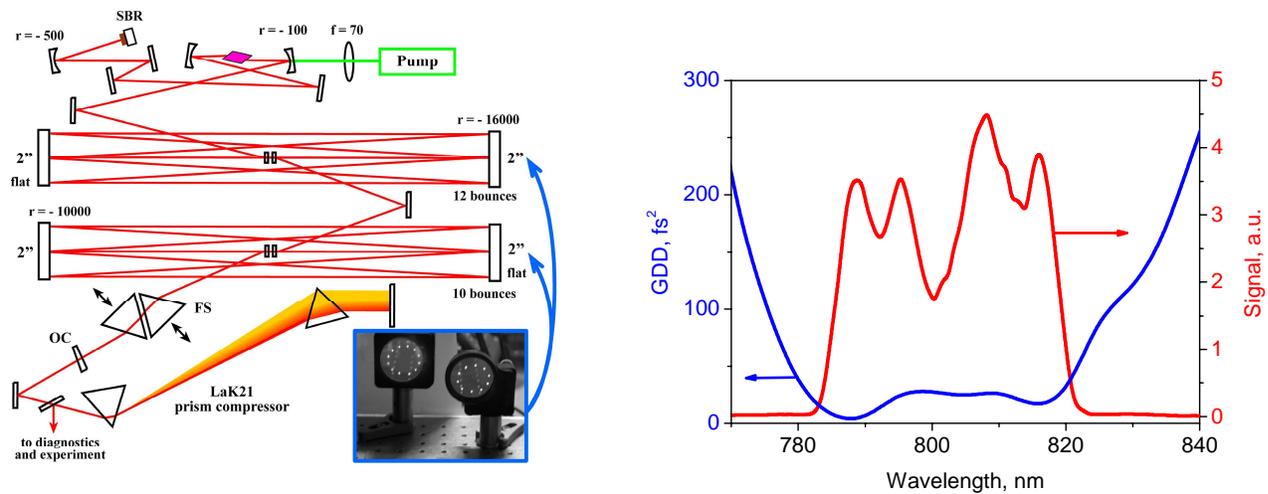


Fig.13. Left: Layout of 500 nJ Ti:sapphire oscillator operating at 2 MHz repetition rate. Right: the generated spectrum (red) and the net-cavity GDD (blue).

Another, even more demanding application of such a smooth-dispersion mirror, is so-called enhancement cavities. They allow storing the pulse energy from a femtosecond oscillators and thus enhance it several orders of magnitude. High harmonics generation in a gas medium embedded into such a cavity has been recently successfully demonstrated<sup>14,15</sup>. The next demands have to be fulfilled in such type of a cavity: first, zero net-cavity dispersion in a range of the spectrum of the femtosecond pulse (770-820 nm in<sup>14</sup>); second, the cavity finesse must be high (~2500 in<sup>14</sup> for high reflectors). The finesse  $F$  of a Fabry-Perot cavity is proportional to  $F \sim R(1 - R)^{-2}$ , where  $R$  is the effective reflectivity of the cavity mirror. The open question is whether similar finesse can be provided by chirped mirrors. If the answer is “yes”, it opens a way to 100  $\mu$ J-level pulses inside such compact cavities operating at MHz repetition rate (seeded with a high-energy oscillator). A big variety of nonlinear experiments in physics can be pursued in this case.

## 5. CONCLUSION

We described 3 types of laser mirrors produced by Helios sputtering system. The design and the realized mirror characteristics (transmission and GDD) have been compared. The measurement data is in good agreement with the design.

The IC3 and CM3 mirrors are useful for broadband Ti:Sapphire oscillators. The reflectivity of IC3 and CM3 are >99% over a broadband range 650-1150 nm. The obtained GDD accuracy is 5 fs<sup>2</sup> and 10 nm in spectral domain.

The LCO mirror is useful for both high-energy long-cavity Ti:Sapphire oscillators and enhancement cavities. They have low losses and high-controlled GDD.

We have shown CMs with high reflectivity. The obtained reflectivity is as high as 99.9% in the wavelength range 720 - 880 nm. The reflectivity of a realized simple quarter wave Bragg reflector is 99.99% in the wavelength range 160 nm around 800 nm.

The production of chirped mirrors is one of the most complex problems in thin film optics. We show that the magnetron sputtering technology with a plasma/ion assisted can be successfully applied for manufacturing very specific, highly-demanding CMs. Still some problems have to be solved in the way to the production of high-quality broadband low-loss chirped mirrors (film stress, damage threshold etc.).

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