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Research Paper

Quality Assessment of Variable Reflectivity Laser Mirrors with High Laser –Induced Damage Threshold and Structural Analysis

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Abstract:
We report the results of an experimental study on the fabrication of a multilayer variable reflectivity laser mirror and the morphological analysis of optical thin film layers. In a design procedure, a series of variable reflectivity laser mirrors are fabricated and characterized with different methods. In this research, based on pulsed solid-state Nd:YAG lasers at 1064 nm, two types of variable reflectivity laser mirrors with (HfO_2/SiO_2) and (ZrO_2/SiO_2) materials are fabricated by electron beam evaporation method. The central reflectivity of R_0 =30 and 35 % for two super Gaussian orders of n=3 and 4 is manufactured and qualified by ellipsometry and laser beam scan methods. The Laser-induced damage threshold experiments demonstrated that, the VRMs with multilayers of HfO_2/SiO_2, have a higher Laser-induced damage threshold than ZrO_2/SiO_2, which was 10 J/cm^2 and 7 J/cm^2, correspondingly. Also, in a novel and exact FESEM approach, the morphological structure of the VRM thin layers was monitored which gives a deeper insight into the layer structure and helped us to calibrate the PVD device for shaped VRM.

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1. INTRODUCTION

High-energy laser pulses, usually extract from unstable laser resonators with large mode volumes that fill the active laser medium. Different methods exist to fully extract the stored energy from laser gain medium, such as telescopic resonators and hard edge mirrors. Besides this advantage, however, there are some drawbacks [1]. The damage threshold of laser mirrors is one of the drawbacks of telescopic resonators for high-energy laser resonators. Non-uniformity in transverse laser beam profile is a problem at the hard edge approaches. Between different methods, unstable laser resonators with Variable Reflectivity Mirrors (VRM) are a modified method that is a good tradeoff between high energy extraction and beam quality. Another advantage is Gaussian modes with large mode volumes and high focusability laser beams. Different approaches have been proposed in the past for implementing of VRMs [2-6]. Several multi-layer types and masking techniques can be used for the manufacture of VRMs [7]. The simplest VRM is fabricated by deposition of a single-variable-thickness layer through a mask onto an antireflection-coated substrate. The peak reflectance will be increased by additional layers. One of the most important aspects of each optical thin film coating, is to characterize the laser Induced Damage Threshold (LIDT) of coated layers. ZrO_2 is an important material for optical thin film deposition and also in the form of nanoparticles has many unique specifications [8]. HfO2/SiO2 layer arrangements, is a suitable choice for high LIDT laser applications and are addressed in many works [9-1٤]. A damage morphology analysis was carried out by the Liu group [9]. They experimentally investigated the damage morphology change process of HfO2/SiO2 reflective film that was illuminated by laser nanosecond pulses at 1064nm. A simulation study was carried out for high reflective film using HfO2/SiO2 design and the thermo-mechanical relationships were evaluated [10].

Recently, the periodic multilayer HfO2/SiO2 for different laser applications is investigated, that demonstrates the efficient operation of such layers for high-power laser applications in the nanosecond and sub-nanosecond lasers [1٥-1٨]. Moreover, the effect of annealing on the optical properties was investigated [1٩].

Though, the first VRM fabrications refer to some years ago, there are only a few reports on morphological and structural studies of VRM laser mirrors. In this research, the design and fabrication approaches of variable reflectivity laser mirrors with (HfO2/SiO2) and (ZrO2/SiO2) materials for a pulsed solid-state Nd:YAG lasers, will be presented. Also, other measurement techniques for layer thickness measurement will be presented. The LIDT of two different coating materials will be evaluated. In this work, in a novel approach, we will try to show that, the morphological study of VRMs during the fabrication, will be an exact method for microscopic verification of the thickness of deposited layers and calibration of PVD devices.

2. DESIGN PROCEDURE

The VRM mirror design is directly related to the laser resonator design. The reflection peak and the shape of the VRM, strongly depend on the resonator parameters that must be carefully optimized to create a good balance between the effective filling of the laser-active material and the diffraction in its aperture. To reduce the reflection of the output mirror, the simplest method, at least analytically, is to create a Gaussian reflection profile. However, the Gaussian curve has a long tail, and the beam spot size must be relatively small to avoid diffraction effects caused by the aperture of the active material. As a result, it seems that flatter Top-hat profiles provide better results. For this purpose, we have considered the reflection profiles with super Gaussian intensity, which are analytically defined below.

$$R = R_0 \exp\left[-2(r/\omega)^n\right] \tag{1}$$

where r is the radial coordinate, R_0 is the peak reflectivity at the center, ω is the mirror spot size or radial distance at which the reflectivity falls to l/e^2 of its peak value, and n is the order of the Gaussian profile. The normalized reflectance profiles for several values of super Gaussian orders n, are shown in Fig.1.

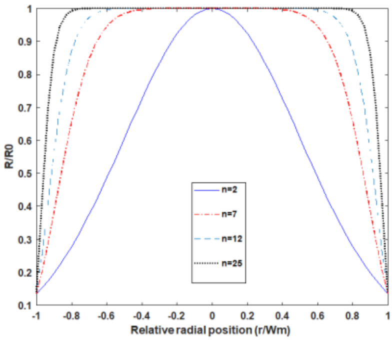

Fig. 1. Intensity reflectance profiles for different Super-Gaussian orders.

The profiles are flatter in the center and reach zero more steeply than the Gaussian curve. Note that if n goes to infinity, the mirror becomes an ordinary uniform profile mirror with radius ω.

3. DEPOSITION PROCESS AND LAYER DESIGN

To fabricate variable reflectivity mirrors, it is better to use materials with a high laser-induced damage threshold, such as SiO_2, ZrO_2, HfO_2 and Ta_2O_5, and substrates such as BK-7, and Fused quartz. To create a layer with variable thickness, it is necessary to use the shadowing effect of a mask in the optical coating process. A schematic design of the general arrangement for VRM production is shown in Figure 2. In this Figure, D, H and h are the aperture diameter, distance between the aperture and substrate, and distance between the evaporation source and aperture, correspondingly.

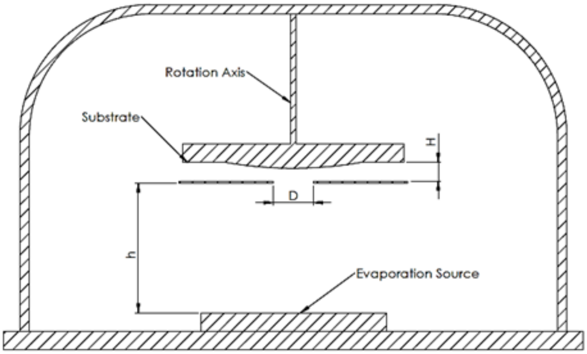

Fig. 2. The experimental arrangement for VRM production

Creating variable thickness to produce VRM mirrors by the methods of 1) variable single layer deposition, 2) variable single-layer deposition between a stack of layers with uniform thickness, and 3) full-layer deposition with variable thickness is carried out [6]. Due to similar changes in thickness layers and continuous deposition process, as well as reducing the time of the deposition process and reducing the risk of contamination, the third method was chosen to make the VRM mirror.

In this work, a fixed mask-fixed substrate with a circular aperture between the evaporation source and the substrate, where the distance from the mask to the substrate is much shorter than the distance from the mask to the substrate, has been used. The optical layer deposition was carried out by a BALZERS BAK 760 evaporation system. According to Fig. 3, the shadowing effect of the mask creates a layer of variable radial thickness, that its characteristics are mainly related to the diameter of the mask (D) and the distance between the mask and the substrate (H). The mask aperture effect creates a thickness that is maximum in the central axis and decreases with radial distance. By controlling the geometrical parameters (D, H), it is possible to control the characteristics of the thickness of the layers and, as a result, the radial dependence of the mirror reflection [4].

The aperture size is comparable with the mask-substrate distance and, consequently, the aperture must be considered as an extended source. It was demonstrated that, there is a cos θ dependence for the angular distribution of evaporated molecules [5]. The geometric thickness of the deposited material at a point of the substrate surface located at the radial distance r, is calculated as follows:

$$t(r) = \frac{4m}{\mu\pi^2 D^2} \int_0^{\frac{D}{2}} \int_0^{2\pi} \frac{H^2 \rho d\rho d\varphi}{(H^2 + r^2 + \rho^2 - 2r\rho\cos\varphi)^2} \tag{2}$$

Where μ is the density of the deposited material, m is the total mass emitted from the source, and (ρ, φ) are the polar coordinates in the aperture plane. Figure 3 shows an example of layer thickness calculations based on distance r and parameters (D, H) [5]. The curves imply that, for a fixed value of D=4mm, by decreasing the H, the deposited layer tends to Flat- Top layer profiles.

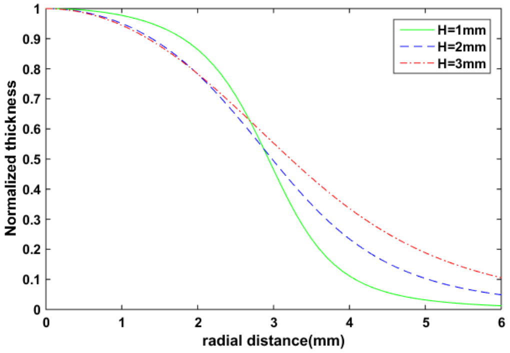

Fig. 3. Dependence of the variable thickness to the radius for D=4mm

In this research, the simulations of layers are carried out by the Essential Macleod layer deposition design software at 1064 nm wavelength. The simulation results for two different center reflectivity are shown in Figure 4. The red (dashed) and black (dot line) curves have 35% and 30% center reflectivity, correspondingly. In this figure, for further references, we named them M#1 for 35% and M#2 for 30% reflectivity. More details of the stacked layers and experimental parameters for M#1 is shown in Table 1(the parameters for M#2 is the same as M#1, but with different layer thicknesses).

Table 1
The details of stacked layers

MATERIAL	ZrO2	SiO2	ZrO2
Layer thickness(nm)	167.2	209	167.2
Partial oxygen Pressure (torr)	1.1×10^{-4}	1.5×10^{-4}	1.1×10^{-4}
deposition rate (nm/s)	0.3	0.5	0.3
substrate temperature(^{0} C)	250	250	250
H (mm)	1	1	1
Refractive index	2.1	1.5	2.1

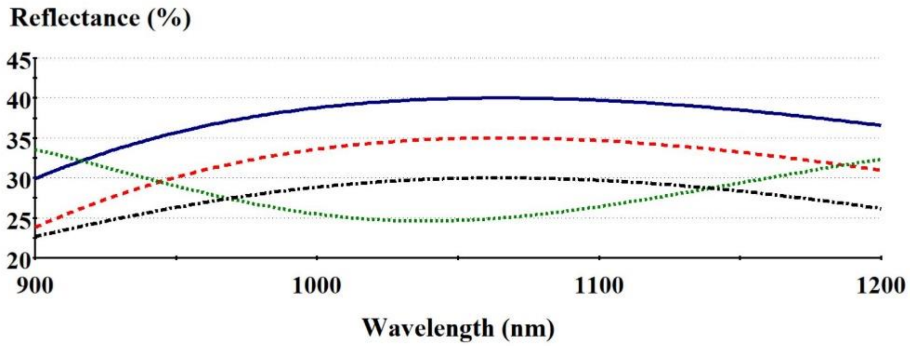

Fig. 4. Different reflectance profile designs with the Essential Macleod software. Red (dashed) and black (dot line) curves illustrate 35% and 30% center reflectivity, correspondingly.

In this work, the e-beam evaporation method, based on physical vapor deposition (PVD) methods, was used. Also, before the masking process and to have an AR- coated substrate, a three -layer of tantalum-oxide (Ta_2O_5) and silicon oxide (SiO_2) alternately were deposited on substrates. In the next step, these substrates were placed in the oven and heated during a program with a specific schedule. The substrate preparation has important impact on the deposited thin film quality [٢٠]. Then, to fabricate the radially varying laser mirrors (VRM), the AR coated substrates was installed in the PVD chamber of Figure 2. The layer deposition was carried out with (SiO_2/HfO_2) and (SiO_2/ZrO_2) materials with three layers alternately and at the accumulation rate of 0.3 and 0.5 nm/s, respectively. It should be noted that the purity of all materials was 99.99%

4. MEASUREMENTS AND VRM CHARACTERIZATIONS

After the design process and mirror fabrication at different conditions, it was necessary to evaluate the various parameters of fabricated VRM mirrors with different methods. In this work, the VRMs was characterized by laser interferometer, laser beam scan, ellipsometry, and FESEM devices.

4.1. ZYGO MEASUREMENTS

First, to ensure the deposition of the layers with radially variable thickness and Gaussian shape, mirror M#1 was placed in the ZYGO laser interferometry device. As illustrated in Figure 5, the final form of coated layers has a Gaussian shape distribution that is in good agreement with our theoretical predictions. The ZYGO device could expose a full view of the VRM layers, but it could not reveal more information about the coated layers. Next, to have more details on morphology and the quality of the layers, we performed more experiments and measurements.

```
08-JUL-1921/15:24:36
Part ID   :
Serial #  :
Analysis  : phase
F/NO      : plano    Fast : off
Averages  : 1        Trim : 0
Calibrate: ON        AGC  : ON
Wave Out  : 0.6328   Scale: 0.50
Reference: none
Remove    : TLT

PV  : 1.061    PTS : 11807
RMS : 0.221
```

Fig. 5. VRM layer profile measured by laser interferometry device

4.2. ELLIPSOMETRY TECHNIQUE

In the next step of our work, for exact measurements of the deposited layers and comparison with simulation results, the ellipsometry technique was utilized. Two types of produced VRMs named M#1 and M#2 with different manufacturing process was selected. The M#1 and M#2 have H=1 and 2.5mm, correspondingly. Table 2 shows the results of measurement and simulations. The layer differences will be related to the masking process. The other error source, maybe associated with the ellipsometry device that is unable to measure the curved sandwiched layers.

Table 2
The ellipsometry measurements of mirror thicknesses for M#1 and M#2 at different H values

MATERIAL	M #1			M #2		
Layer	ZrO_2	SiO_2	ZrO_2	ZrO_2	SiO_2	ZrO_2
Simulated thickness	167.2	209	167.2	167.2	209	167.2
Measured thickness	135.5	202.3	144.9	131.8	180	109.5

4.3. LASER BEAM SCAN

In this set of experiments, to characterize the mirror reflection profile and the comparison between theoretical and experimental methods, the VRM mirrors were installed in a profile metric arrangement depicted in Figure 6. The laser source is a pulsed Nd:YAG laser system that works in the free running mode at 1064nm, 150 μs pulse duration, and 1Hz pulse repetition rates. The laser output beam that works as a probe beam in this experiment, is reduced to less than 100 µm by a Plano-Convex lens with a 50mm focal length.

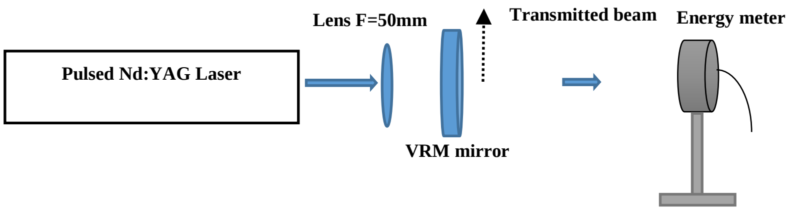

Fig. 6. The experimental arrangement for optical coating characterization by laser beam scan

Then, the reflection profile of M#1 and M#2 was obtained based on the scanned points of Figure 6. As can be understood from Figure 7, there is a good compromise between theoretical simulations and experimental values. Figures 7a and 7b illustrate the measured and fitted curves for two different super-Gaussian laser mirrors. Also, the details of measured parameters is presented in Table 3.

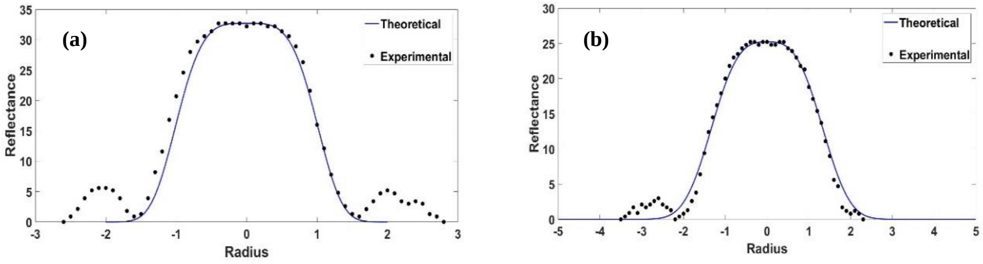

Fig. 7. Reflection profiles obtained from VRM mirrors with super- Gaussian orders of : a. n=4 for M#1 and b. n=3 for M#2

As demonstrated in Table 2, two mirrors, named M#1 and M#2 were fabricated during our optical coating processes. The results are in good agreement with Macleod simulations of Figure 2. Also, it is shown in Table 3, that there is some error for R_0 between the design and measured values. The error sources can be related to optical coating devices or measurement tolerances. Moreover, the errors can be related to the difference between the design and measured values of optical coatings. It should be noted that, in our subsequent coating processes, these errors were reduced significantly. Also, it is clear that, by increasing the H values, the super Gaussian order (n) decreases.

Table 3
Different parameters of produced VRMs

MIRROR NUMBER	H(mm)	n	ω_m(mm)	R_0	R_0 Error
M#1	1	4	1.4	32%	~10%
M#2	2.5	3	1.8	26%	~8.6%

5. LASER TEST

As mentioned in the introduction, the aim of this work is the fabrication of VRM mirrors for pulsed Nd:YAG lasers. In the subsequent experiment, to validate of mirrors operation, one of the mirrors, M#1, was installed in a pulsed solid state unstable laser resonator as an output coupler and operated at free running and Q switch modes with an unstable Cassegrian resonator with the magnification of M=1.3. The extracted energy was about 480mJ at 7ns pulse duration, which shows about 1.2% optical to electrical extraction efficiency. Also, for the evaluation of super Gaussian behavior and transverse mode structure, the laser beam profile was recorded by a laser beam profiler. The laser beam profile is shown in Figure 8. Using the above procedures, we could fabricate the rectangular shaped VRM for solid-state lasers with slab geometry (inset right of Figure 8. b).

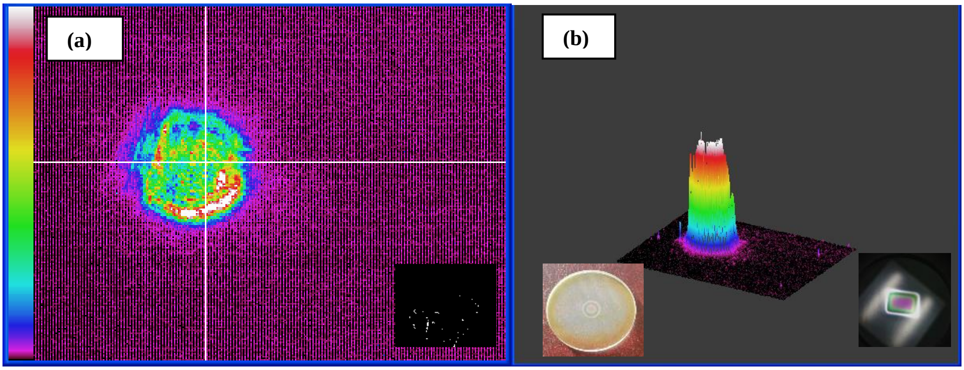

Fig. 8. (a) The near-field (inset: laser spot trace on laser-sensitive paper) and (b) far-field profile of the laser beam for M#1 with circular symmetry (inset left: circular symmetry fabricated VRM, inset right: rectangular shape fabricated VRM)

6. MORPHOLOGICAL STUDY

For a deep study of the coating process and characterization of the optical thin film layers, a morphological analysis was carried out by a Field Emission Scanning Electron Microscopy (FESEM) system. FESEM is one of the most common techniques used to characterize surface morphology. Because of the special form of shaped layers at VRM, the usual thickness measurement methods will be incorrect. The FESEM morphological study will be an efficient offline method to layer thickness control.

In this step, another VRM sample, named M#3, was investigated by a FESEM device. The multi-layer VRM laser mirror was designed in the form of a

$ZrO_2/SiO_2/ ZrO_2$ arrangement, with a thickness of 209.14/170.32/196.07, correspondingly. The mirror was designed to have a central reflectivity R_0= 30% and the super Gaussian order of n=3. The Macleod simulation curve is illustrated in Figure 9a. Subsequently, the coating process and necessary measurements were carried out. The FESEM surveillance, helped us to calibrate the PVD device for favorite layer thicknesses and, consequently the desired R_0. Finally, after a few optimizations and FESEM checking, the desired VRM was fabricated. The last FESEM picture of M#3 is shown in Figure. 9b. Comparison between simulation and FESEM results show that, there are slight differences between simulation and FESEM measurements. One of the advantages of morphological study, is the verification of a PVD device operation and to insertion of a suitable correction factor to the coating process and device calibration.

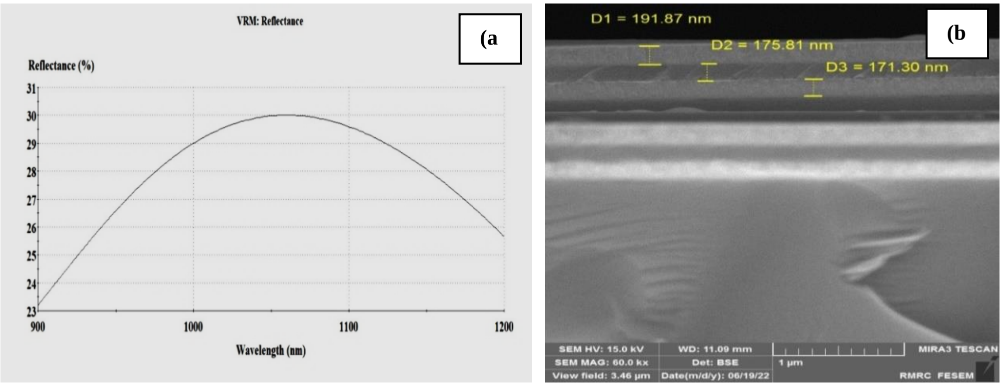

Fig. 9. (a) The Macleod simulation results (b) the FESEM scan of the deposited layer of M#3

7. LIDT EXPERIMENTS

Laser-Induced Damage Threshold (LIDT) is one of the most essential parameters for a laser mirror. This parameter will be critical for high-power laser system design.

To measure the laser-induced damage threshold, two samples of VRM mirrors, including ZrO_2/SiO_2 (M#4) and HfO_2/SiO_2 (M#5) materials, were investigated in an LIDT setup. The LIDT setup includes a pulsed Nd:YAG laser cavity with a 7 mm laser rod that works at 10 Hz pulse repetition rates. The laser operates at a Q-switched mode of operation at 7ns pulse duration. Also, the laser energy was 500mJ, which was focused by a suitable Plano -Convex lens with a 250mm focal length.

Next, by installing the produced mirrors in the LIDT setup, the mirrors M#4 and M#5 were tested by 30 successive pulsed. The results of LIDT experiments are shown in Figure 9. The results demonstrate that the VRM mirrors made of ZrO_2/SiO_2 (M#4) were destroyed by irradiating pulses with an energy density of 7 J/cm^2. But the mirrors made of HfO_2/SiO_2 (M#5) have higher LIDT than M#4 and were damaged at 10 J/cm^2 laser energy density. The microscopic images of damaged samples are shown in Figure 11. The experimental results of LIDT confirms that, the fabricated multilayer HfO_2/SiO_2 partial reflectors, are suitable for high energy laser oscillators with VRM output couplers.

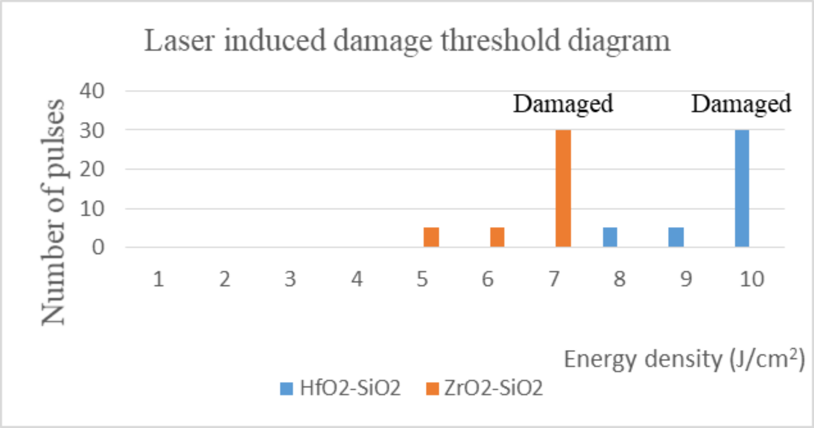

Fig. 10. The laser-induced damage threshold diagrams

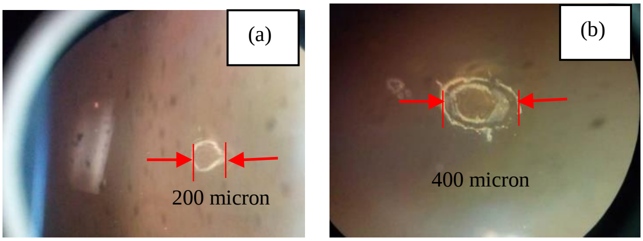

Fig. 11. Laser-induced damage on (a) HfO_2/SiO_2 / HfO_2 and (b) ZrO_2/SiO_2 / ZrO_2 VRM samples

8. CONCLUSION

In conclusion, the results of the simulation and fabrication of a VRM laser mirror for 1064nm solid-state lasers are presented. The results of exact practical measurements with different methods show that there is a good compromise between simulation and experimental approaches. The performance of these mirrors in Q-switched solid state laser resonators, shows the necessary durability against unwanted optical damage. We concluded that the laser mirrors with HfO_2/SiO_2 structure, have more LIDT than ZrO_2/SiO_2 VRM mirrors and can withstand nanosecond laser pulses more than 10 J/cm^2. In this research, the qualification of a shaped VRM by the FESEM measurements was carried out. Also, it was concluded that, the morphological study of deposited layers, will be an exact method to calibrate the optical coating devices.

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