

Laser damage of Silica and Hafnia thin films made with different deposition technologies

Laurent Gallais, Jérémie Capoulade, Jean-Yves Natoli, Mireille Commandré, Michel Cathelinaud, Cian Koc, Michel Lequime

Institut Fresnel (UMR CNRS 6133) - Université Paul Cezanne - Université de Provence - Ecole Centrale Marseille

Domaine Universitaire de St Jérôme - 13397 Marseille Cedex 20 - France

ABSTRACT

A comparative study is made on the laser damage resistance of monolayers coatings made with different technologies. HfO_2 and SiO_2 thin films have been deposited on fused silica substrates with Dual Ion Beam Sputtering, Electron Beam Deposition (with and without Ion Assistance) and Reactive Low Voltage Ion Plating technologies. The laser damage thresholds of these coatings have been determined at 1064nm and 355nm using a nanosecond pulsed YAG laser, and a 1-on-1 test procedure.

1. INTRODUCTION

HfO_2 and SiO_2 are widely used materials for the production of laser damage resistant multilayer coatings.¹⁻³ From numerous studies, it appears that e-beam-deposited HfO_2/SiO_2 multilayer coatings shows higher damage threshold than the same coatings deposited with energetic deposition techniques.⁴⁻⁸ But these last techniques have better optical, mechanical and environmental properties. The improvement of the laser damage resistance of dense layers has therefore a considerable interest for the community. In this context we report in this paper the recent development made at the Fresnel Institute in the development of dense coatings for high power applications.

This paper is a summary of works published in reference⁹ for SiO_2 and¹⁰ for HfO_2 . Then one should refer to these publications for more details.

2. MATERIALS AND METHODS

2.1 Samples description

The substrate choice and preparation (cleaning and polishing) is critical for the production of coatings for high power applications and it can ultimately limit the thresholds for a given coating.^{11,12} For this study, we used 1-inch diameter fused silica substrates polished for high power applications. Herasil was used for the silica coatings and Corning 7980 was used for hafnia coatings. For each coating material (either silica or hafnia) all the substrates (either Herasil or Corning 7980) were coming from the same batch and have been polished at the same time.

To clean these substrates we used an automatic aqueous cleaning procedure, involving ultrasonic immersion and detergents followed by de-ionized water rinsing and drying.

The aim of the study was to test samples at 355 and 1064nm. However, to have comparable results at a given wavelength, it is necessary to assure that the optical thickness of the different samples are the same, in order to have the same standing electric field in the coating. Then in the case of hafnia, a first serie of samples half wave at 1064nm has been made for testing at 1064nm and a second serie, half wave at 355nm, was made for 355nm testing.

In the case of silica however the optical thickness of the layer is not a relevant parameter as regards the electric field distribution, because of the very low refractive index difference between the substrate and the layer. Then the same samples were used for 1064 and 355nm tests.

Further author information: (Send correspondence to L. Gallais)
E-mail: laurent.gallais@fresnel.fr, Telephone: 33 4 91 28 80 72

2.2 Deposition process

The SiO_2 coatings were deposited by the following techniques: Electron Beam Deposition, Ion Assisted Deposition, Reactive Low Voltage Ion Plating (RLVIP) and Dual Ion Beam Sputtering.

The hafnia samples were prepared by Electron Beam Deposition using either a Hafnium or Hafnia source (respectively the samples will be called EBD-Hf or EBD-HfO₂ in the following text), Reactive Low Voltage Ion Plating and Dual Ion Beam Sputtering (DIBS).

The process parameters used to produce these coatings are reported in table 1 and 2.

Table 1. Process parameters for SiO_2 coatings

Procedure	Deposition technique			
	EBD	EBD-IAD	RLVIP	DIBS
Starting material	Silicon 99.999%	Silica	Silicon 99.999%	Silicon
hline Base pressure		3×10^{-7} mbar		7×10^{-7} mbar
Chamber pressure	9×10^{-4} mbar (O ₂)	3×10^{-4} (O ₂)	9.5×10^{-4} mbar (Ar+O ₂)	3×10^{-4} mbar (Ar+O ₂)
Deposition rate	0.8nm/s	0.4nm/s	0.8nm/s	0.1nm/s
Substrate temperature	250°C no heater	50°C no heater	150°C no heater	50°C no heater
Anode voltage				

Table 2. Process parameters for HfO_2 coatings

Procedure	Deposition technique			
	EBD-Hf	EBD-HfO ₂	RLVIP	DIBS
Starting material	Hafnium 99.5% Umicore granulate 1-10mm	HfO ₂ tablets Merck Patinal	Hafnium 99.5% Umicore granulate 1-10mm	Hafnium 99.4% Neyco
liner	carbon	molybdenium	carbon	
evaporation gun	10kV	10kV		1kV, 400mA acceleration : 350V
Assistance parameters			plasma source arc current and voltage : 55A, 66V Anode voltage : 42V	Ion gun : 375V, 350mA acceleration : 300V
Base pressure	3×10^{-7} mbar	3×10^{-7} mbar	3×10^{-7} mbar	7×10^{-7} mbar
Chamber pressure	5×10^{-4} mbar (O ₂)	5×10^{-4} mbar (O ₂)	6×10^{-4} mbar (Ar+O ₂)	5.5×10^{-4} mbar (Ar+O ₂)
Deposition rate	0.9nm/s	0.9nm/s	0.2nm/s	0.05nm/s
Substrate temperature	250°C lamp heaters	250°C lamp heater	200-250°C no heater	50°C no heater
Anode voltage			42V	

2.3 Laser damage tests

The configuration used for LIDT measurement is described on figure 1. The laser source is an injected Nd:YAG laser (Quintel YG 980) with a pulse duration of 11 ns (effective pulse duration at $1/e^{13}$) and a maximum repetition rate of 10 Hz. The laser can operate at 1064nm or 355nm, with a maximum energy of 1.5 J at 1064 nm and 450mJ at 355nm. It works at a repetition rate of 10Hz to keep a good stability of energy and beam characteristics. A mechanical shutter permits to work in single shot mode. Energy of the beam is controlled with a variable attenuator (half wave plate and polarizer).

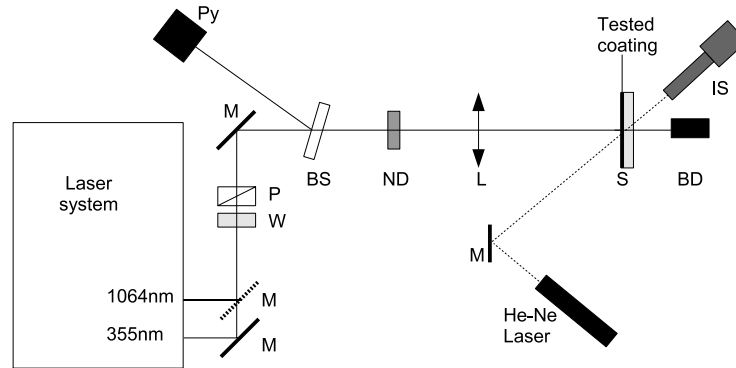


Figure 1. Experimental configuration for laser damage tests

The laser beam (linearly polarized and in normal incidence) is focused on the front face of the coated sample (see figure 1) with a plano-convex lens at 1064nm ($f=35\text{mm}$) and an objective in the case of 355nm ($f=11.5\text{mm}$). The spot size has been measured in the sample plane with a beam analyser : the spot diameter is $44\mu\text{m}$ at 1064nm and $3\mu\text{m}$ at 355nm (diameter at $1/e$).

The damage detection is done by comparing the area before and after irradiation with an imaging system (magnification : X216) and an image processing software. The damage criterion is then any visible modification detected with this system.

For each shot, energy is measured with a pyroelectric detector and recorded. The fluence is then calculated according to the ISO standard by using the effective spot size.¹³

The LIDT measurement setup is fully automated, which allows to measure laser damage probabilities with a great number of points. In this study, the damage test procedure 1-on-1 is used : by counting the number of damaged regions at each fluence F we estimate and plot the damage probability curve $P(F)$. To have a good accuracy of the measurement, each curve $P(F)$ is plotted with 1000 data points that involve 20 different fluences and 50 tested regions at each fluence.

3. RESULTS AND DISCUSSION

The test results are plotted on figures 2 (355nm) and 3 (1064nm) for silica, and figures 4 (355nm) and 5 (1064nm) for hafnia. The plain lines curves on these figures are the fit of the experimental points, made with a statistical model relying on the hypothesis of defect initiation (see ref¹⁴). The shape and threshold of these curves are linked to the initiator characteristics (density, nature,...). However the analysis of these curves is not discussed in details in this paper since our study is more focused on the comparisons of the technologies.

One difficulty in laser damage testing is often to compare laser damage performances of a particular material between different testing facilities (large discrepancies are found between publications). Indeed the damage threshold can be linked to many factors such as the test procedure, the beam profile, the wavelength, the pulse profile and duration, the number of shots... For this reason we also give in the following sections the LIDT of the substrates (polished, cleaned and stocked with the coatings substrates) and tested in the same experimental conditions.

3.1 Silica

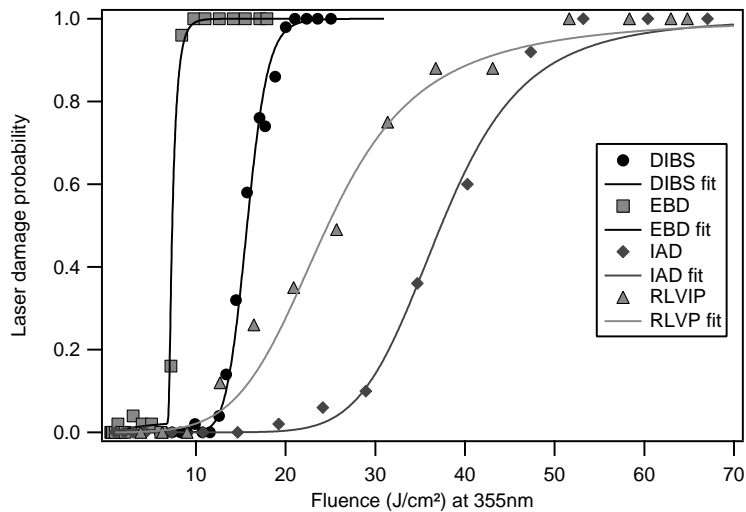


Figure 2. Laser damage probability curves measured at 355nm for the different silica samples

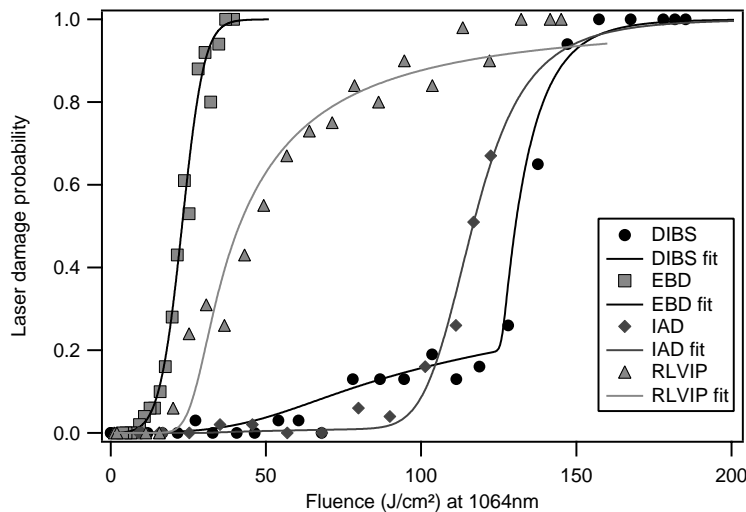


Figure 3. Laser damage probability curves measured at 1064nm for the different silica samples

The LIDT of the films, i.e. highest fluence where no damage is observed, are the following :

- EBD : $4\text{J}/\text{cm}^2$ at 355nm and $12\text{J}/\text{cm}^2$ at 1064nm
- EBD-IAD : $19\text{J}/\text{cm}^2$ at 355nm and $30\text{J}/\text{cm}^2$ at 1064nm
- RLVIP : $10\text{J}/\text{cm}^2$ at 355nm and $20\text{J}/\text{cm}^2$ at 1064nm
- DIBS : $9\text{J}/\text{cm}^2$ at 355nm and $35\text{J}/\text{cm}^2$ at 1064nm
- Substrate : $30\text{J}/\text{cm}^2$ at 355nm and $70\text{J}/\text{cm}^2$ at 1064nm

High laser damage threshold is often reported in the literature for low density thin films, such as those obtained by Sol-Gel or Electron Beam Deposition.^{1,3,6,15} In our study however, we observe that the dense layers (obtained with ion-assisted process) exhibit a very good threshold in the infrared (DIBS) and the UV (IAD)

compared with layers manufactured by classical EBD. This indicates that a good quality in terms of defects has been obtained for these dense layers. Finally, the Dual Ion Beam Sputtering technique appears to be a promising technique for the production of high laser damage threshold coatings, since it also leads to very good mechanical, structural and optical properties for the layers.

However it is known that it is relatively easy to obtain silica layers with a high LIDT, and that the high index material in a multilayer stack will be the limiting material.

3.2 Hafnia

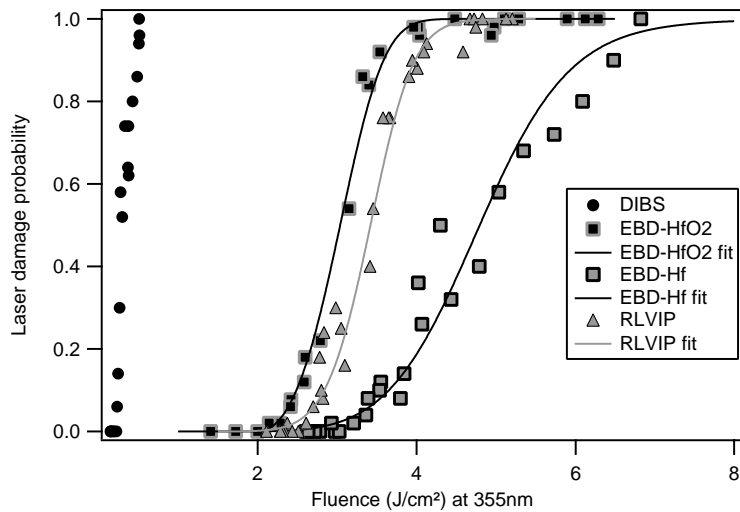


Figure 4. Laser damage probability curves measured at 355nm for the different hafnia samples

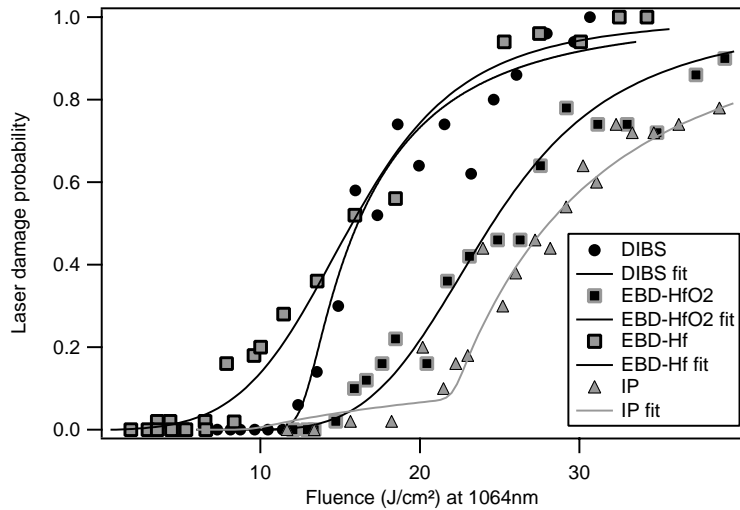


Figure 5. Laser damage probability curves measured at 1064nm for the different hafnia samples

The LIDT of the films, i.e. highest fluence where no damage is observed, are the following :

- EBD-HfO₂ : 2.1J/cm² at 355nm and 14.5J/cm² at 1064nm
- EBD-Hf : 2.8J/cm² at 355nm and 3.5J/cm² at 1064nm
- RLVIP : 2.3J/cm² at 355nm and 15.5J/cm² at 1064nm

- DIBS : 0.22J/cm² at 355nm and 12J/cm² at 1064nm
- Substrate : 18J/cm² at 355nm and 70J/cm² at 1064nm

The best threshold in the IR is found for the RLVIP technique and the worst result is obtained for EBD-Hf. This is quite surprising compared to general results in this field where EBD-Hf is found to be the best technology, particularly at 1064nm. This indicates that the deposition parameters chosen are far from optimal for the EBD-Hf process and can be further optimized for 1064nm applications (a high roughness is also found for these films). The results for the RLVIP coatings are interesting and promising since no data were published to our knowledge for this technique.

For the DIBS thin films tested at 355nm, a very low LIDT is found. This is associated to a high extinction coefficient and a damage morphology evidencing burning and melting of the film. In this case, damage is linked to the high intrinsic absorption level of the film and thermal melting of the material is the main laser damage mechanism. This can be due to a stoichiometric problem of the film, such as oxygen deficiency, linked to the bombardment process or the relatively low substrate temperature compared to other technologies. The optimization of the DIBS process for UV applications seems delicate : no report of high LIDT DIBS films in the UV are published to our knowledge.

4. CONCLUSION

Single layer of SiO₂ and HfO₂ have been deposited on fused silica substrates with different state of the art assisted and non-assisted deposition techniques. The coatings have been laser-damage tested at 355 and 1064nm (12ns). The LIDT were found to be strongly dependent on the deposition technique but also on the test wavelength : no correlation were found between LIDT at 355 and 1064nm. High laser damage threshold were found for dense coatings. This results appear promising for the production of laser damage resistant multilayer coatings.

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Q I saw in your chart the deposition rate for the ion beam is .05 nm per second. This rate would mean one hour or two for deposition, correct?

A It depends on the number of layers.

Q Ion beam deposition is obviously a practical technique, but you were your rates realistic at it being so slow?

A Yes, it was very slow, but if you want a high density threshold, it will be a problem.

Q You did some computations of absorption, by the hafnium metallic particles and I wondered what kind of dielectric functions you used?

A Yes, hafnium. The reference values for the reference for hafnium which is made up of metallic crystals were used.

Q What I saw there was data for two sizes of metallic clusters, but not for continuously changing sizes. Did you do some extrapolations?

A We have taken data for the big sizes. It's not the defects sizes of one nanometer between the damage sites, but or a large difference.