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Defect geometries and laser-induced damage in multilayer coatings

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ABSTRACT

A correlation between laser-induced damage and the height of a coating defect was found from an examination of $\text{HfO}_2/\text{SiO}_2$ mirrors made by three different coating vendors. The nodular defects in these reactive e-beam deposited mirrors were studied using the combination of SEM, optical microscopy, FIB and AFM techniques. Each vendor had small defects in common, but characteristically different large defects. Also the majority of seeds that caused the defects were made of hafnia, not silica. The apparent mechanical stability of the defects within the coating plays a major role in the laser resistance (1064 nm and 10ns) of a high damage threshold coating.

1. INTRODUCTION

The design of high power lasers for fusion is currently limited by optical coatings. These coatings are multilayer interference coatings used as turning mirrors and polarizers. The limiting aspect of these coatings is their resistance to high power laser illumination and can be associated to the microstructure of the films, more specifically, nodular defects in the film.^{1,2,3} These nodules grow as a result of self-shadowing effects initiated at seed particles either present on the substrate or deposited on the coating during multilayer deposition. The defect height, h , and diameter, D , and their relationship to seed depth, T , and seed diameter, d , are defined by

$$D = \sqrt{cdT} \quad (1)$$

and $h=d$ and c is a constant.⁴ In the present study, focused ion beam cross-sectioning techniques were used to better characterize the defect geometries and the seed particles.

It is generally observed that all defects observed in a coating do not damage at the same laser fluence. In order to identify any correlations between damage susceptibility and defect geometry, atomic force microscopy (AFM) has been used to characterize individual coating defects prior to laser damage experiments. Earlier results for coatings from Vendor A showed that the damage susceptibility of the defects increased with the height of the defect, with a significant increase in susceptibility for defects with heights greater than $0.6 \mu\text{m}$.⁵ The correlation of damage threshold to defect diameter was less clear and was attributed to the dependence of defect diameter on defect height predicted by the simple model for nodule shape (Eqn. 1).

The purpose of the present paper is to determine the generality of these observations, at least for e-beam $\text{HfO}_2/\text{SiO}_2$ coatings. Reported here are the results of the characterization and laser damage studies of coating defects supplied by two additional vendors, B and C. The long term goal of this work is to understand the initiation and growth mechanisms of defects in optical coatings, suggest methods for the minimization of defects, and gain an understanding of the failure mechanisms that limit the capabilities of a coating.

2. DEFECT CHARACTERIZATION

Optical microscopy, AFM, scanning electron microscopy (SEM), and focused ion beam (FIB) techniques were combined in the following fashion to detail the microstructure of coatings and defects. To characterize individual defects, a sample was marked with a reference fiducial. The sample was first observed with a Nomarski light microscope to find an area with a high density of defects. The sample was then coated with a thin layer of metal to make the surface conductive for imaging in a high resolution SEM. The defects were next scanned with an AFM to measure the physical shape of the defects in three dimensions. The sample was then placed in a FIB system and the defects were cross-sectioned. After cross-sectioning, the defects were again

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imaged with the high resolution SEM for subsurface geometry measurement of the nodules. Images produced by this final step provided the quantitative observations that follow for samples of e-beam deposited hafnia/silica multilayers.

2.1 The "classic" nodule

The defect shown in Fig. 1a is classified as a "classic" nodule because it most closely matches the general appearance of a nodule as described earlier by Eqn. 1. The nodule is cylindrically symmetric, i.e., in top view it is circular. The transition region from the nodule's dome to the mirror's flat surface is smooth and continuous. Another major topographical feature of the classic nodule is that the general microstructure of the coating on the nodule's surface is indistinguishable from that of the surrounding surface. The structure of the boundary region between the defect and the surrounding layers changes as the deposition progresses. Close to the seed, this boundary region is convoluted and the individual layers are not continuous. But, as more material is deposited, the boundary heals and the layers become continuous. The figure shows the shape of the seed, its depth, and the shape of the layers near the seed. The micrograph shows that the seed is close to spherical as expected for a classic nodule.

Assuming that the seed is produced by the ejection of coating material, its composition might be deduced from the contrast in the SEM image. The silica layers appear as the darker material in the image because the silica is less conducting than the hafnia. The half-wave over-coat of silica is clearly visible in the image. The brighter layers then must be the hafnia. The seed shows similar contrast to the hafnia layers and can therefore be assumed to be hafnia. Auger analysis, presented previously by this group, confirms this assumption. Note that the seed was deposited at the end of the third layer of hafnia. Such information can be important to the operator of the coating apparatus, because it might be correlated to a particular deposition event that causes seed ejection.

2.2 The "non-classic" nodule

The term "non-classic" nodule is used to describe the defect in Fig. 1b because it retains some of the properties of the "classic" while losing others. The nodule is cylindrically symmetric, i.e., in top view it is circular, like the "classic." However, in this case, the edges of the nodule are very rough and the transition is rough from the nodule to the surface. In addition, the coating surface dips next to the edge of the nodule, which is most likely caused by the larger seed dimensions shadowing the incoming material next to the defect. As with the "classic" nodule, the general microstructure of the coating on and off the nodule is very similar.

The main difference between the "classic" and "non-classic," however, is that the seed in Fig. 1b is larger than that in Fig. 1a by 2.5 times which leads to the extremely convoluted boundary region next to this nodule. It also appears to have voids in this region (voids are basically indistinguishable from insulators to the SEM). This profile view shows how the coating dips next to the defect and, in this case, the entire boundary is a convoluted region. This seed was deposited some time after the third layer of the coating run and it again appears to be a hafnia seed.

2.3 Vendor comparison

The coatings studied were deposited by three vendors with extensive experience in high-damage-threshold coating deposition. Some characteristics of the deposition chambers and the coatings are given in Table 1. In general, the deposition tank sizes, deposition angles, e-guns, and source material preparation were different for each vendor. For example, coatings from vendors A and B were deposited in deposition tanks capable of coating large substrates (1-m diam.), whereas vendor C used a standard size (2' x 2' x 3') box coater. Damage tests on the coatings showed that they all displayed laser conditioning effects and had similar damage thresholds.⁶

Defect height vs. diameter plots for vendors B and C are shown in Fig. 2 (See refs. [3] and [5] for vendor A). Over half the defects seen in each sample had heights $<0.5\text{ }\mu\text{m}$, corresponding to seeds of the same diameter. If all seeds were found on the substrate or were deposited at the same time, the height-width data would fall onto a single curve. The spread in the data in Figs. 2a and 2b, however, indicates that the seeds were deposited over a range of depths.

Table 1: Some characteristics of the deposition chamber and respective coatings

Vendor	Source-to-substrate distance, cm	Damage threshold, J/cm ² (3 ns, 1064 nm)		Predominant defect types
		S:1	R:1	
A*	>48"	15	20	classic, semi-classic
B	>48"	8	27	classic, multi-lobe
C	~18"	13	26	classic, spatter, pits

* Data reported in refs. [3] and [5]

For both vendors B and C, the small defects with heights $<0.4 \mu\text{m}$ showed classic structures such as those in Fig. 1a. For larger defects the more complicated multi-lobe or "spatter" defects shown in Fig. 3, were often observed. These larger defects are generally associated with complex seed shapes.

Vendor C appears to have fewer large defects. We found, however, many defect craters on this sample. In general, the craters are due to the natural ejection of large, mechanically unstable nodules. More work needs to be done to characterize these crater defects.

3. LASER-INDUCED DAMAGE

3.1 Experimental Procedure

Laser illumination was performed using standard laser-damage testing techniques using a 1064-nm Nd:YAG laser with a 10-ns pulselength. At the damage test plane a $1/e^2$ beam diameter of 1.6 mm was obtained by focusing the beam with a 3-m lens. Laser fluences were determined through beam profiling and total energy measurements.

Defect characterization and damage detection were done using a Large Area AFM (Digital Instruments, Inc.) including a computer controlled sample positioning stage. The system allowed rapid and accurate movement of the sample between a high resolution ($\sim 850\times$) optical microscope, the AFM head, and the laser illumination area. Defects found using the optical microscope were repositioned under the AFM for characterization, moved under the laser for illumination, and then moved back to the AFM to determine if damage had occurred.

The coatings studied were deposited on 2" diameter BK-7 substrates. The coatings were high reflectors of quarter-wave stacks designed for use at normal incidence. The top layer of the coatings were half-wave SiO_2 overlayers. The surfaces were cleaned by drag wiping with alcohol prior to testing.

3.2 Laser Damage Results

Each defect characterized was also illuminated with a single laser pulse of a predetermined fluence. Figure 4 plots the height of each defect vs. the fluences with which it was illuminated. The dark squares show cases when damage occurred. The lines indicate apparent transitions between regions of no damage, occasional damage, and consistent damage. For both samples, the damage probability increased with defect height. While the exact nature of the transition zone between low and high damage probability is different for samples B and C, as well as sample A, the general increase in damage susceptibility seems to consistently occur over a defect height range near $0.5 \mu\text{m}$. The transition to high damage probability at relatively low defect heights for sample C might be attributed to the easily ejected spatter defects found on this sample.

Figure 5 plots illumination fluence vs. defect width for the same set of defects shown in Fig. 4. As with defect height, there seems to be a general increase in damage probability as the defect width increases. The correlation is not as clear as for defect height, and, based on the results for studies on sample A, the dependence is believed to be associated with the dependence of the defect width on the seed diameter (i.e., defect height) and depth.

For each sample, a few crater defects, formed by the intrinsic ejection of nodules from the coating (due to stress or other mechanical instabilities), were illuminated. As in the case for sample A, these crater defects did not appear to show an increased damage susceptibility over the surrounding film.

4. DISCUSSION AND CONCLUSIONS

The results of site specific damage experiments on defects in $\text{HfO}_2/\text{SiO}_2$ coatings have consistently shown that laser damage susceptibility can be correlated with defect geometry. While the exact nature of the increase in damage susceptibility with defect height varies between vendors, there is typically a strong increase in damage susceptibility as the defect height increases over 0.5 μm . The defects with these large heights are initiated by large and often complex seeds. The resultant defects often have complex boundaries resulting in poor mechanical stability. It is therefore not surprising that these defects often damage first.

We must also consider, however, that the laser-induced ejection of these weakly held large defects may not be detrimental to the coating. Since the resulting craters are the same size as the original nodule, this process may not be easily detected by optical microscopy. The laser-induced ejection of the more strongly held, typically, more classic defects may cause more damage to the surrounding coating when they finally fail. This collateral damage may then propagate upon further illumination. Therefore, while the larger defects may show the highest damage susceptibility, we should not assume that the smaller defects are acceptable for high fluence applications.

Laser conditioning is believed to be related to the mechanical stabilization of defects. The low fluence illumination may mechanically alter or stabilize the defects so that subsequent high fluence illumination will not cause catastrophic damage. Some of the more mechanically unstable defects may even be ejected from the coating during conditioning.

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6. AUSPICES

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Figure 1. SEM micrographs of hafnia/silica multilayers, a) small spherical seed produced "classic" nodule, b) large spherical seed produced non-ideal nodule.

Vendor B

(a)

Vendor C

(b)

Fig. 2

Fig 3

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4. APPLICATION TO THREE VENDORS

Figure 3 AFM images of characteristic defects for three vendors, a) vendor A has non-ideal defects created by liquid ejecta from the source, b) vendor B has defects caused by solid particulates, and c) vendor C has defects like B that are initiated by solid seeds.

(a)

(b)

Fig. 4

(a)

(b)

Fig. 5

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