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Effect of Silica Overlayers on Laser Damage of HfO_2-SiO_2 56° Incidence High Reflectors

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ABSTRACT

A series of hafnia/silica, oblique incidence (56°), 1064 nm high reflectors (HRs) were prepared and coated with silica overlayers of varying optical thickness from $\lambda/2$ to 4λ in order to determine the effect of an overlayer on the laser-damage resistance of the HRs. The stress and laser damage thresholds for S and P polarization of the HRs were measured, and the damage sites for P polarization examined by Atomic Force Microscopy (AFM). All the multilayers were found to be in compression, with an intrinsic stress increasing with overlayer thickness. The presence of an overlayer and its thickness did not affect the damage threshold significantly. However, the presence of an overlayer greatly influenced the size and morphology of the damage. First, the overlayer prevented catastrophic "burns" of the hafnia top layer. Second, as the overlayer thickness increased, two distinct damage morphologies were found: jagged pits and round craters. The diameter of these pits and craters then increased somewhat with thicker overlayers. The depths of the pits and craters also increased with overlayer thickness, and the depths showed failure occurring at the interfaces below the hafnia layers. The side-wall angles of the craters were shallower with thicker overlayers, but there was no angle dependence for the pits. The craters showed fracture-like features and a small hillock or pit on their bottom surfaces. No correlation of damage morphology to conditioning or fluence was found.

1. INTRODUCTION

The addition of a silica overcoat has long been thought[1,2] to improve laser damage performance of quarter-wave high reflectors (HRs) of various types, though little systematic work has been done to document the improvement or to relate it to the thickness of the overlayer. Wu et al.[3,4] reported that for normal incidence tests on single silica layers with 10 ns-1.06 µm laser pulses, the layers showed damage with no dependence on film thickness. On the other hand, the damage threshold of ZrO_2/ SiO_2 and TiO_2/ SiO_2 reflectors showed a very strong dependence on overlayer thickness (see Fig. 1). By measuring the scattering loss and observing the microstructure, Wu et al. concluded that

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variations of the overcoat thickness lead to changes in overcoat's surface morphology and interface structure. Samples with thicker overlayers tend to have smoother top surfaces and finer microstructures. But when the overcoats are too thick, stress-induced cracks appear on the surface and the damage threshold drops.

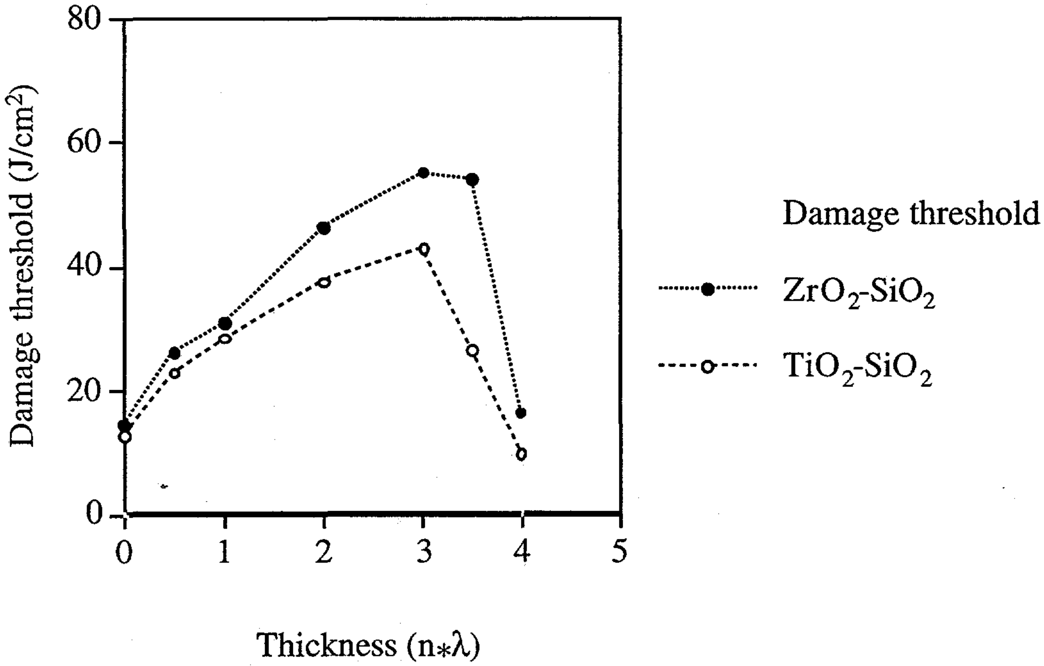

Fig. 1. Damage thresholds variation with the silica overlayer thickness for titania/silica and zirconia/silica multilayers (from Wu et al.[4]).

Recent laser interaction studies conducted in parallel with this study by Sheehan et al. for 45°-incidence, 1064 nm wavelength, hafnia-silica mirrors[5] have shown craters similar to those seen in this study. Sheehan found that delamination occurred close to the surface and occurred preferentially at interfaces. Usually, the overcoat and the top hafnia layer, or the overcoat, the top hafnia layer and one layer pair were ejected, and a rim around the hole was formed. The present study is consistent with these results but a more quantitative evaluation is made here.

In this study, we have attempted to determine whether HfO_2/ SiO_2 HRs with silica overlayers would show a thickness dependence of the damage threshold like that seen by Wu et al.. In addition, the effects of the overlayer on the damage morphology and on the stress in the multilayer were investigated. Since the damage threshold is consistently lower for P polarization, understanding the P polarization damage is more critical to improving mirror performance, so these sites were investigated in detail. The samples in this study were conditioned under ramped (R:1) and constant (S:1) fluence in different areas, and damage sites from both areas were examined. Lastly, this study was conducted at oblique incidence (56°) so the electric field distribution was not axisymmetric and was different from that in Wu's test.

This article will first report the stress measurements and damage threshold measurements in S and P polarization. It will then present a detailed AFM analysis of the morphology of the damage sites for P polarization and describe how the defect diameter, depth, and side-wall angle varied with overlayer thickness. Conclusions will include implications for further research in explaining the mechanism of the damage observed. The results show substantial differences from Wu's results: within experimental error, the damage threshold did not depend on overlayer thickness for the HRs studied.

2. EXPERIMENTAL

Hafnia/silica multilayers HRs were prepared by reactive e-beam evaporation. The hafnia was evaporated from a metal Hf source and the silica from pure SiO_2. The chamber was evacuated to a base pressure of 5 x 10^{-7} torr then the coating run was performed at partial pressures of 2 x 10^{-4} torr of Hf + O_2 and 5 x 10^{-5} torr of SiO_2 + O_2. The substrate temperature was maintained at 190°C and the deposition rates were about 2Å/s for HfO_2 and 5Å/s for SiO_2. Coating thickness was monitored *in situ* by an optical monitor, switching materials by shuttering when the quarter-wave condition was met. All the HRs were prepared during the same coating run. Thin quartz samples were simultaneously coated to measure stress; these measurements were done using interferometry for the lower stress values and using a mechanical stylus for higher stress values.

The optical multilayer was designed to reflect at an angle of 56° and wavelength, λ, of 1064 nm. The stack was deposited following the sequence:

Substrate -- $(H\,L)^9$ H -- (m • L)

where H is a hafnia layer, 156 nm thick (quarter-wave optical thickness), L is a silica layer, 224 nm thick, and m is either 0, 2, 4, 8 or 16, giving overlayers of optical thickness 0, λ/2, λ, 2λ and 4λ.

The samples were then subjected to standard laser damage threshold tests with 3 ns pulses; 60 seconds exposure at a shot frequency of 10 Hz and at 1064 nm wavelength. Sites were subjected to constant (S:1) and ramped (R:1) laser fluence and tested at S and P polarization to measure the damage thresholds. Damage sites of various fluences were examined by a long-scan-range AFM (Digital Instruments). Damage site dimensions were measured directly from the AFM scans.

3. RESULTS

The laser damage thresholds are summarized in Fig. 2. The damage thresholds of the overcoated HRs were not significantly affected (i.e. showed no trend outside experimental error) by the thickness of the overlayer. The non-overcoated samples tested 13.4 J/cm^2 in P polarization but 19.6 J/cm^2 in S polarization.

The intrinsic (or growth) stress is shown in Fig. 3 as a function of overlayer thickness. No direct correlation could be made between the overall stress and the laser damage performance. Measurements of stress[6] at room temperature indicated that the hafnia layers were in tension while the silica layers were under compression.

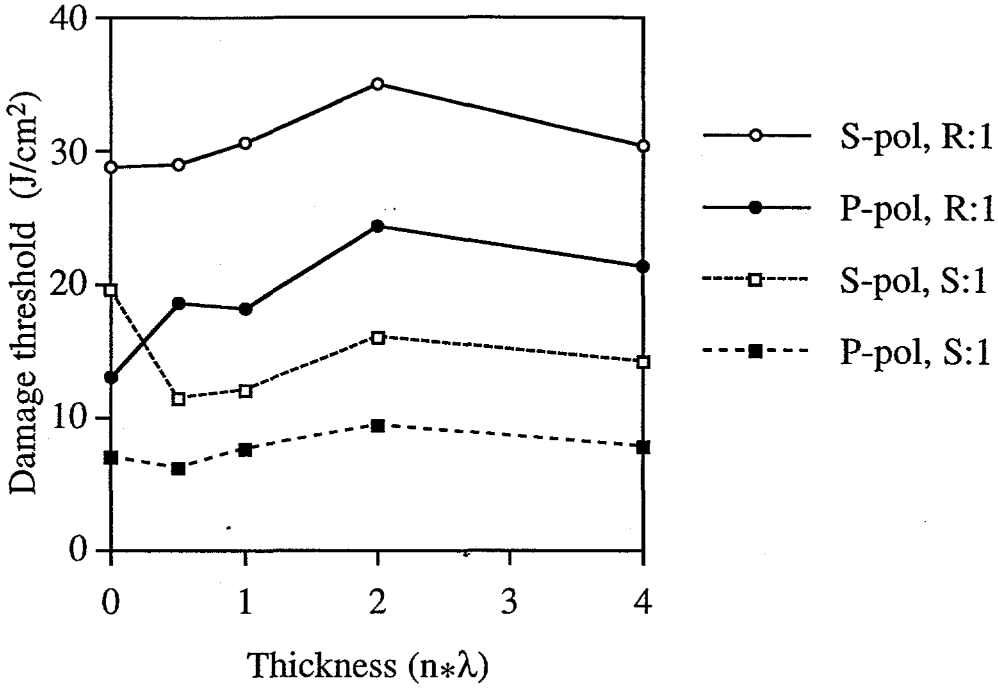

Fig. 2. Damage thresholds variation with the silica overlayer thickness, for S:1 and R:1 conditioning and S and P polarization.

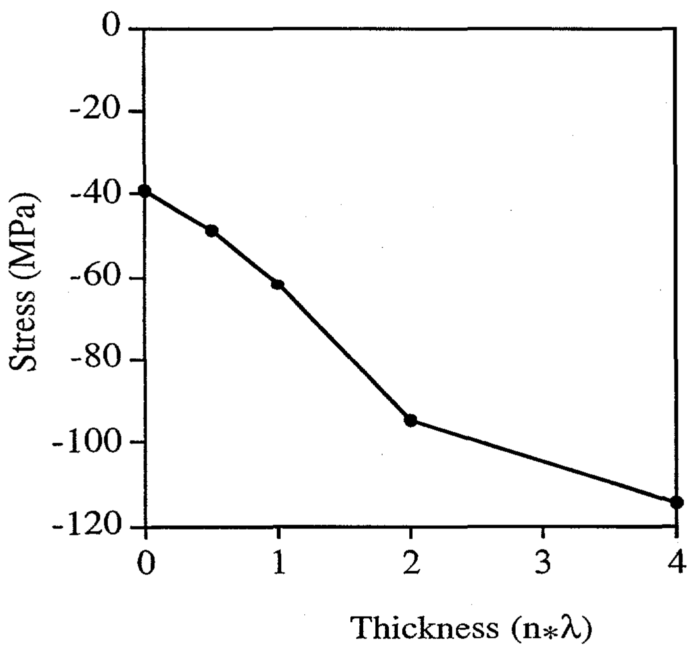

Fig. 3. Intrinsic stress variation with silica overlayer thickness.

Damage occurred in two distinct morphologies, differing in all three parameters measured: damage-site diameter, depth, and side-wall angle. The two morphologies also differed consistently in shape: one type was jagged and the other smooth and round. The round sites (craters) were almost always flat-bottomed; they had a very definite depth and often had a smaller pit with concentric ring-like features on the bottom surface. The jagged sites (pits) had various shapes at their bottoms. A typical pit and a typical crater are shown in Fig. 4 for an HR with a 4λ overcoat. The statistics for the occurrence and geometry of the two types are presented in the next two paragraphs.

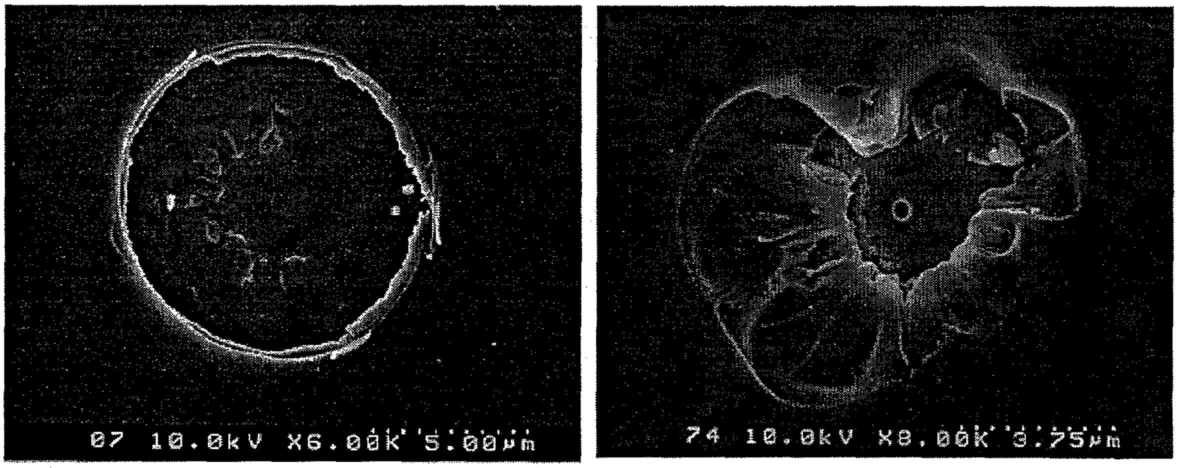

Fig. 4. Typical crater *(a)* and pit *(b)* damage sites of an HR with a 4λ overlayer after laser illumination in P polarization S:1 at a fluence of 10.1 J/cm^2.

Histograms of the defect diameters and depths are shown in Fig. 5, as a function of overlayer thickness. The open symbols are for the pits and the filled symbols for the craters. This figure presents the entire data set collected and every effort was made to survey all the defect types present. Superimposed on the depth data are the geometrical thickness of the overlayer and the quarter-wave layers beneath, as calculated from their optical thickness, the incident angle, and the indices of refraction (1.45 for SiO_2 and 1.89 for HfO_2 at 1064 nm).

A plot of the side-wall angles as a function of overlayer thickness appears in Fig. 6. To measure this angle, a line was drawn, on a line-out from the AFM scan, from the upper rim where the pit wall meets the film surface to the lower rim where it meets the flat bottom of the pit, or to the deepest point if the bottom was not flat. The angle was then measured between this line and the horizontal film surface. This measurement was done at eight points spaced equally around the circumference of the pit and the values averaged. In many cases the fracture surface changed angle as it passed through layers of different materials and sometimes had ledges parallel to the film surface. This is the cause of the noise in these data.

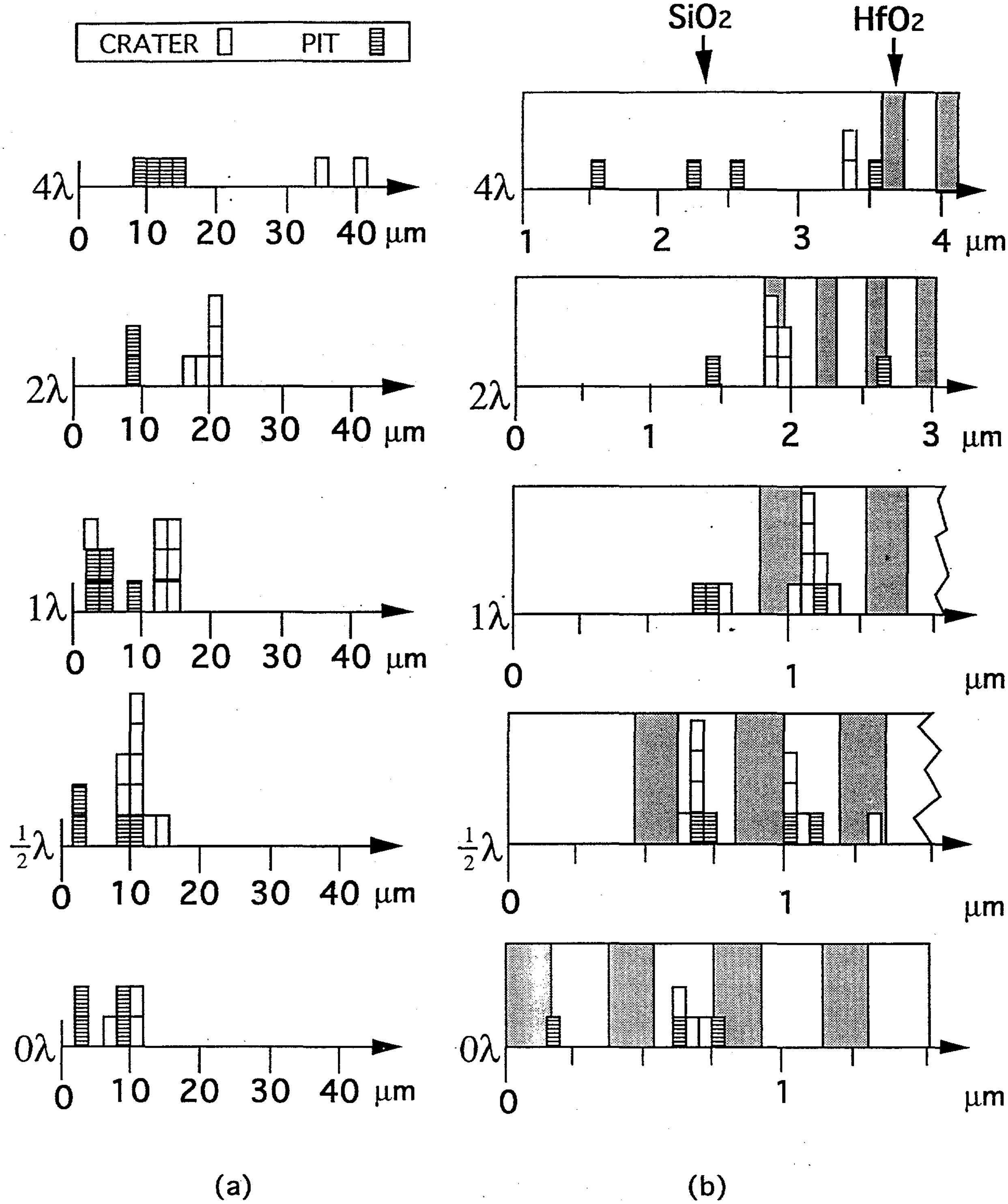

Fig. 5. *(a)*Histogram of pit and crater diameters for overlayer thickness 0 to 4λ. *(b)* Histogram of pit and crater depths, with cross-section of HR layers for reference. Horizontal scale in *(b)* is adjusted for 2λ and 4λ because of the thick overlayer.

4. DISCUSSION

The sites examined were conditioned before damage testing. To include the greatest possible number of damage sites, sites from both S:1 and R:1 conditioning were examined. No correlation of the geometry of the damage sites to the prior conditioning was found; the damage in the histograms is randomly distributed between S:1 and R:1 sites.

The pit width data indicate that there are two clearly distinct damage types - the widths clump around two values (near 9 µm and 20 µm for the 2λ overlayer, for example) and these clumps correlate well with the pit and crater morphologies. Furthermore the difference in diameters and the correlation with pit shape lessen with decreasing overlayer thickness, and for no overlayer there is no longer any distinction. Thus the overlayer is somehow involved in the failure mechanism and strongly affects the mode of failure.

The interpretation of the pit-wall angle is less clear because of the irregular fracture angle mentioned above. However they and the depth data indicate a fracture mode which is affected by the different properties of the layer materials and shows a definite tendency for failure at interfaces (delamination). The depths clearly cluster about certain values which are separated by approximately one layer-pair thickness, which indicates a preference for failure at one type of interface, in this case those with a HfO_2 layer above and a SiO_2 layer below. However, for very thick overlayers (4λ), fracture seems to typically occur above the first hafnia layer (i.e. within the overlayer itself).

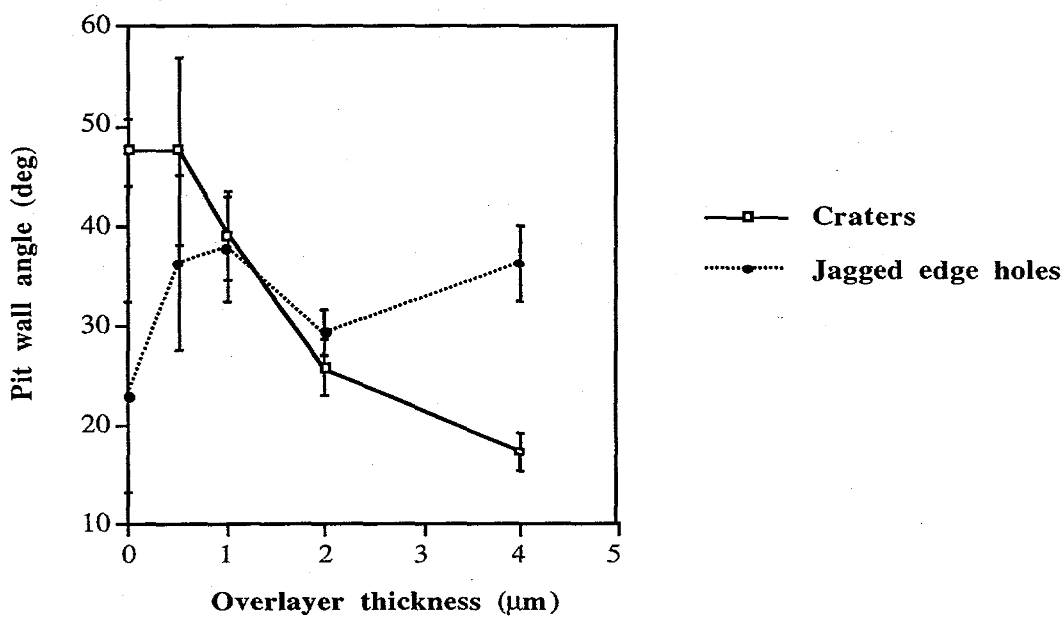

Fig. 6. Pit and crater side-wall angles vs. overlayer thickness.

The damage described covers the range of laser fluences used in the damage test, generally in a range from 50% to 200% of the damage threshold. Within this range, for a given overlayer thickness, the geometry did not vary significantly with fluence. As for change of geometry with overlayer thickness at constant fluence, a subset of data, chosen as close as possible to 20 J/cm^2 over the range of overlayer thickness, showed the same variation of defect geometry with thickness as the whole data set.

The mechanism of this delamination and damage is not clear from what is now known. In the craters, the bottom surface showed in most cases a small secondary pit or a small hillock near the center. It is known that the nodular defects frequently observed[7,8,9] in similar mirrors are particles (seeds) of HfO_2 or SiO_2 embedded during deposition of the multilayer. Since the height of the small secondary pits and hillocks (0.5-2 μm) is similar to the size of the seeds as measured by previous AFM and Focused Ion Beam sectioning studies, it is plausible that the craters result from the ejection of a nodule near the top of the HfO_2 layer, with the fracture surface following the nearby interface for some distance before moving to the surface. The bottoms of the jagged pits showed similar features. It has been suggested[10] that such damage could also occur as a result of heat generation at highly absorbent centers and that subsequent heat radiation by such centers causes delamination along weak links such as interfaces.

The existence of two distinct types of pits suggests either two types of defects or two modes of failure from the same set of defects. The failure mode may be determined by the location of the seed or absorption center within the layer, and the preferential delamination could simply results from the seed's proximity to that interface. In a related study[11] using AFM to target specific defects before the laser pulse and relating them to damage morphologies afterwards, nodules were associated with jagged pits resembling those in this study. Round craters like those in this study were also seen, at locations where no defect had been detected beforehand in the optical scanner of the AFM. Thus the round craters may not result from nodules. It is possible, however, that nodules or other defects too small to be seen in the optical system existed at these sites before illumination.

Such features would be observable, however, if a wide AFM scan of the area were made beforehand. Since several such features could often be found within 300 μm of the targeted nodule, a systematic check of 100 μm x 100 μm area laser illumination could easily cover an area in which one or more craters could later be expected. With such a test it may be possible to associate the craters with some observable surface features in the same way the pits seem to be associated with nodules. This would in turn shed some light on the reason for the two damage morphologies and the difference between them, and may provide information on the relationship of damage to film microstructure and surface roughness.

5. CONCLUSIONS

The experiments reported here have shown that the presence of a silica overlayer improves the stability of the HRs against laser damage. While it can greatly reduce the damage area, it does not substantially influence the value of the damage threshold. Typically, with the presence of an overlayer, the damage site area tends to increase with the thickness of the overlayer. This damage was mostly observed in two morphologies: jagged pits and smooth round craters, which had different diameters, depths and side-wall angles, and showed a different change of these characteristics with the overlayer thickness. This suggests they correspond to two different failure modes, that they may propagate in a different manner, or that they originate from two different types of seeds. Except for very thick overcoats, failure occurred preferentially at the interfaces below the hafnia layers and

occurred near the top of the HR stack. The samples with thick overcoats showed damage within the overcoat itself. No correlation of damage geometry with prior conditioning or fluence was found. Thicker overlayers led to greater intrinsic compressive stress in the HRs that could not be related to the damage threshold.
In conclusion, this study has pointed out the importance of the top layers for oblique-incidence HRs. In particular, it has shown that the damage threshold may be governed by interfacial phenomena occurring in the top few layers, and that damage size and morphology could be influenced by the presence of the overlayer and its thickness.

6. ACKNOWLEDGMENTS

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