Section 2
ADVANCED TECHNOLOGY DEVELOPMENTS

2.A Development of Optical Coatings for the OMEGA Upgrade

An intensive in-house effort in the past year has produced coatings and devices that will survive the peak 351-nm fluence expected in the OMEGA Upgrade. The stringent ultraviolet requirements for the upgrade stem from the decision to place the frequency-conversion crystals in the laser bay (rather than at the target chamber).[1] At that time, only one report of satisfactory laser-damage thresholds for 351-nm high reflectors existed.[2] Reports of satisfactory distributed phase plates (DPP's) did not exist, and tests indicated the thresholds of existing designs would not suffice. Modification of the laser beam-transport-system design (see Section 1.A, this issue) lowered the thresholds from those listed in the Preliminary Design Document.[1] These new thresholds are listed in Table 44.III. As seen in the table, fluences in the blue were lowered for high reflectors and DPP's, but remained the same for antireflection coatings.

The pending need for upgrade coatings dictated the development of improved coating technologies. Since we investigated variations on established methods, issues other than damage, such as scale-up to large apertures, optical performance, and environmental performance, are assumed to be known and previously addressed by the optical community. Optical coating of high reflectors by evaporation is the most common technique used for high-energy lasers. We report on coatings using standard evaporation methods and on those using an ion-assisted deposition (IAD) technique. Sol-gel deposition methods are the preference for antireflection coating.

Table 44.III: Previous and current maximum fluence levels for the OMEGA Upgrade 351-nm components (main pulse, 0.6 ns).

	Previous (Preliminary Design Document)	Current Design	Process
Frequency-conversion crystals	3.8 J/cm^2	3.8 J/cm^2	Sol-gel
Relay lenses	3.8 J/cm^2	3.8 J/cm^2	Sol-gel
Transport mirrors	3.8 J/cm^2	2.9 J/cm^2	Dielectric evaporation
Refocusing asphere	3.6 J/cm^2	2.9 J/cm^2	Sol-gel
Distributed phase plate	3.6 J/cm^2	3.6 J/cm^2	Ion etch/sol-gel

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Development of a sol-gel coating capability and related damage-test results are reported for various substrates. Finally, development of an etching technique for DPP's that can handle substantially higher fluences than previous designs is described.

Development Procedure

The study began by choosing materials and process variables that gave favorable results in the past, then depositing and damage testing the coating. Favorable results were explored further. Generally, no attempt was made to further characterize the coating or understand the reason for the performance. While this approach is not scientifically satisfying, it is a very effective development method when the in-house coating and damage test groups can closely collaborate. Also, since there is no well-established set of precursors to laser damage, testing remains the only reliable measure of the reliability of a coating in a large laser system.

Coatings were deposited on well-characterized, 5-cm-diam substrates of either BK-7, Pyrex®, or fused silica. Each substrate was cleaned using equipment and procedures established for cleaning the large substrates used on OMEGA. The aqueous process uses a series of mechanical scrubs, rinses, and soaks in ultrasonic baths. The final rinse with 18-MΩ water takes place in a class-10 clean-room environment where the substrate is left to dry. The 54-in. and 28-in. deposition chambers used for this study were both equipped with electron-beam evaporation sources for reactive-gas (oxygen) evaporation. With the exception of some of the ion-assisted coatings, all substrates were held at 200°C during deposition. This is based upon in-house experience and an extensive parameter study with tantalum pentoxide,[3] which gave the best damage results for low substrate temperatures. These low temperatures also reduce the possibility of coating fracture. The other deposition parameters were adjusted to give low average absorption for thin films in the 300- to 400-nm range.

All coatings were tested in the LLE damage test facility with 351-nm, 0.7-ns FWHM pulses. Both 1-on-1 and N-on-1 testing were performed on the samples. In 1-on-1 testing (see Fig. 44.33), a new site is chosen for each laser shot. A minimum of ten sites are usually examined. The 1-on-1 damage threshold is defined as the average of the highest nondamaging fluence and the lowest damaging fluence seen in all sites. In N-on-1 testing, one site is subjected to successive laser pulses, each one increasing in fluence above the previous pulse until damage is observed. Three to ten sites are tested in this manner, and the fluences at which damage occurs are averaged to give the N-on-1 damage threshold. N-on-1 testing is more typical of the operation of a large laser facility, demonstrating a hardening effect in the tested surface.[4] Damage is assumed to have occurred whenever a new scatter site appears within a 100 × 100-μm area observed under dark-field incandescent illumination.

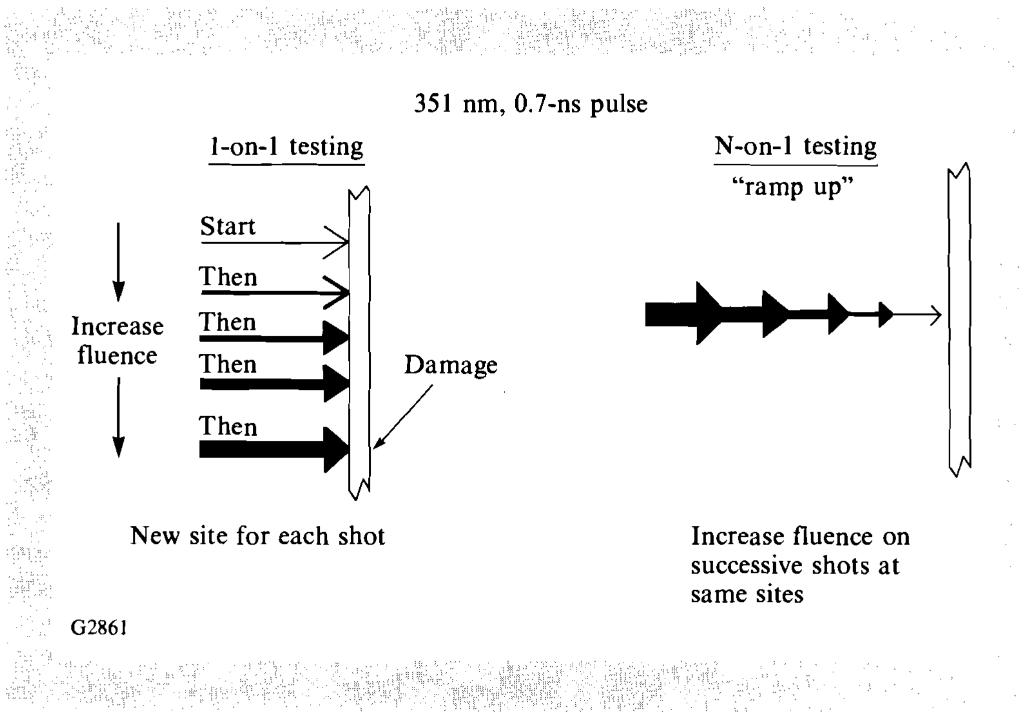

Fig. 44.33
In 1-on-1 testing (left) a site is subjected to laser radiation, examined, then the process is repeated at a higher fluence on a new site until damage is observed on several sites. In N-on-1 testing (right), the fluence is gradually increased at a single site. Between each laser pulse, the surface is examined for damage.

High Reflectors for 351 nm

Two pieces of equipment were used to produce high-reflector coatings: a 54-in.-wide chamber with a planetary-substrate rotation (for high uniformity on large substrates) and a 28-in.-wide system with a single-substrate rotation. The two chambers allowed for two different approaches to achieve high damage thresholds; a materials/design survey was conducted in the larger coater, while the smaller coater concentrated on one pair of materials under different process conditions. The effects of oxygen-ion assist were primarily investigated in the smaller chamber with the best results later repeated in the larger chamber. Typical deposition parameters for some materials are given in Table 44.IV. The results from the materials/design survey are reported first.

Table 44.IV: Deposition parameters for various high-reflector coating materials—200°C substrate temperature.

Material	Deposition Rate Å/s	O_2 Backpressure (Mbar)
SiO_2	5	4×10^{-5}
MgF_2	5	none
Y_2O_3	2	1.2×10^{-4}
Sc_2O_3	1.5	1.7×10^{-4}
ZrO_2	2	1.3×10^{-4}
HfO_2	0.5	1.7×10^{-4}
Ta_2O_5	1–2	1.8×10^{-4}

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Laser-damage thresholds (LDT) of single layers of materials do not correlate well with thresholds of those same materials in multilayer designs.[5] Therefore, we tested only high-reflector designs, choosing materials that had shown potential for high LDT at 351 nm in other studies. All of the coatings were made to meet component specification for the OMEGA Upgrade; blue reflectors were designed to have reflectivity greater than 0.995 for both polarizations at the given incidence angle. Thus, while the coatings may physically differ in the number of layers, they all are functionally equivalent.

The N-on-1 results of the first set of coatings are shown in Fig. 44.34. The threshold is identified by the circle or triangle, and the error bars represent one standard deviation for the measurements on each sample. Yttria/silica designs were considered because they can be stripped from a surface without repolishing.[6] Despite considerable effort, these coatings never achieved acceptable thresholds. Scandia and zirconia-based coatings performed well, as they had in other studies,[2] with scandia/silica coatings

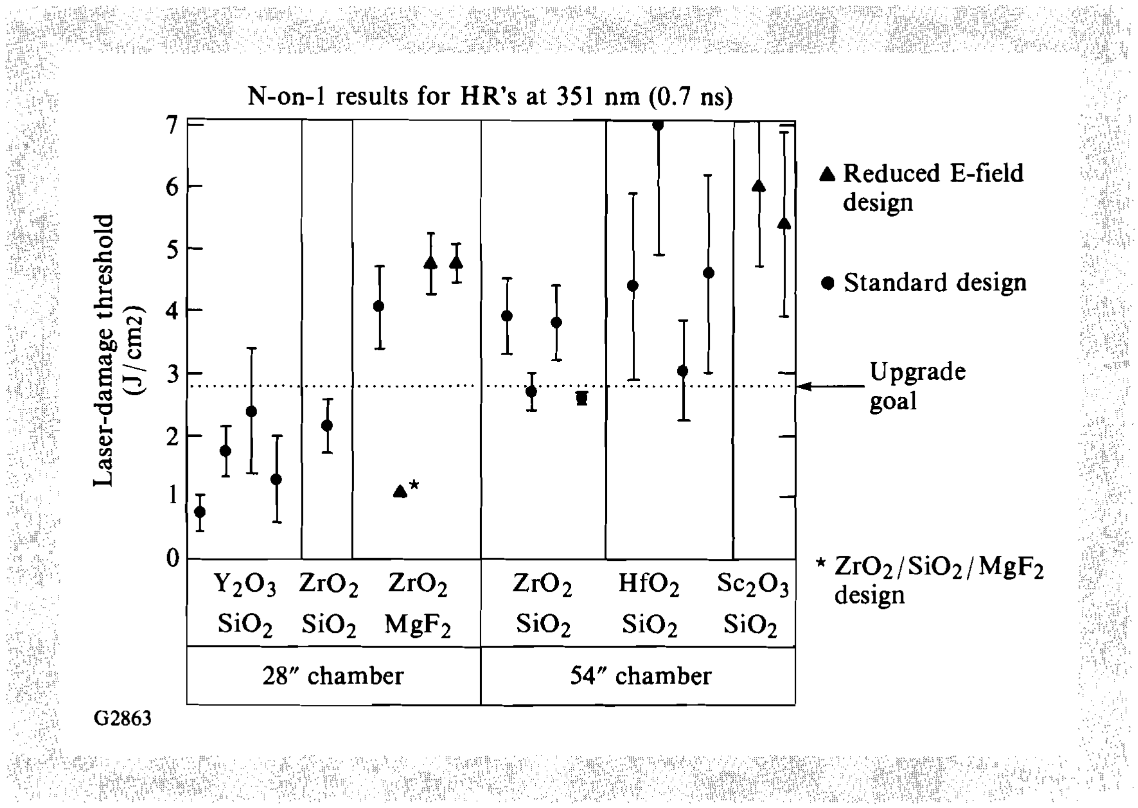

Fig. 44.34
N-on-1 laser damage test (LDT) results for a materials study with evaporated coatings in two different chambers. The design requirement for the OMEGA Upgrade is indicated by the dotted line. The error bars show a one-standard deviation of the data from the mean.

exceeding the upgrade goal considerably. There are several noteworthy results in this data: (1) hafnia/silica coatings performed very well contrary to previous results[2]; (2) zirconia/magnesium fluoride coatings also had high thresholds except for a hybrid material design; (3) the best results came from designs with reduced electric field in the outer four layers; and (4) the data suggest that coatings made in the larger chamber have higher thresholds. The difference seen between the chambers may be attributed to variations in process parameters for the two systems.

Design variations using reduced E-fields and starting materials were investigated for the promising hafnia/silica combination in the 54-in. chamber. Reduced E-field designs of high reflectors[7] have exhibited higher damage thresholds in other studies.[2] In these designs, the time-averaged electric-field intensity is reduced in the most damage-prone material, which is hafnia in the current coatings. The peak electric-field intensity is also moved from the interface into the low-index layer where the damage resistance may be greater. Our data, seen in Fig. 44.35, tends to agree with these premises. The highest thresholds in this study were consistently produced by reduced E-field designs.

At the time of this study, one vendor[8] supplied two grades of hafnia (standard grade and UV grade). Spectrographic analysis showed the UV grade had significantly less titanium and zirconium contamination and less trace contaminants. Despite the lower contamination, Fig. 44.35 shows that for a standard high-reflector (HR) design there was actually a slight decrease in the threshold for the more pure hafnia, while no perceived difference was observed for the reduced E-field designs between the two materials. This was an intriguing result since spectroscopy confirmed that the UV-grade hafnia did have a lower absorption edge, which typically corresponds to a higher laser-damage threshold.

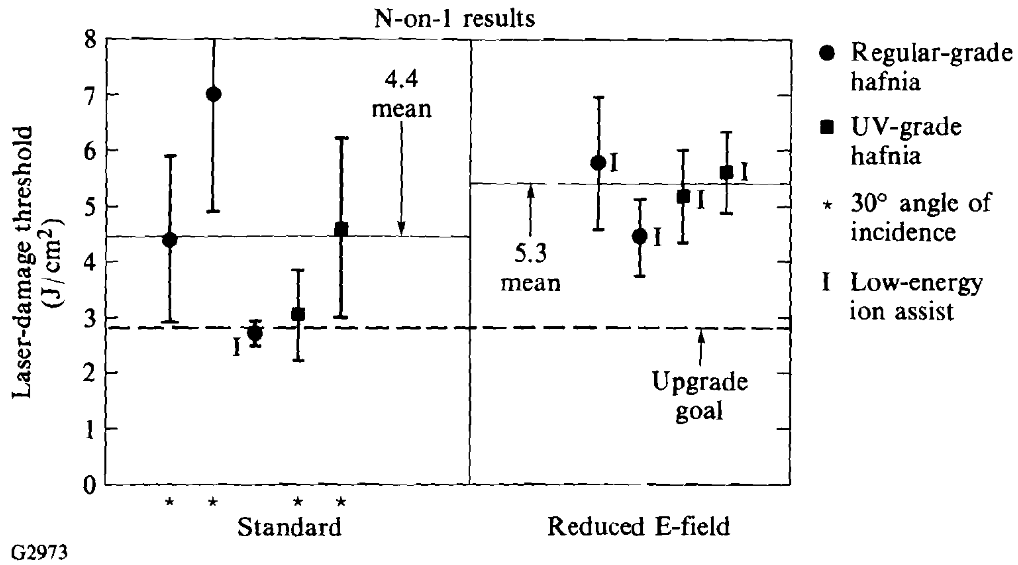

Fig. 44.35
Laser damage test (LDT) results for hafnia-silica high reflectors. The combination of a reduced E-field design and ion assist produced the most consistent results in this study.

Coatings using magnesium fluoride as the low-index material were made with zirconia or hafnia as the high-index material. The zirconia coatings produce the highest thresholds for any HR made in the smaller chamber (Fig. 44.36). Oddly, the same coating made in the larger chamber performed poorly. The standard design with hafnia (UV grade) and magnesium fluoride also performed poorly at about 3 J/cm^2. The same materials in a reduced E-field design almost doubled in damage threshold (5.4 J/cm^2). The small error bar suggests that HfO_2/MgF_2 coatings may perform better than ScO_2/SiO_2 coatings on a laser system. These results show the magnesium-fluoride-based HR coatings are likely candidates for upgrade transport optics, but they require further study and process development.

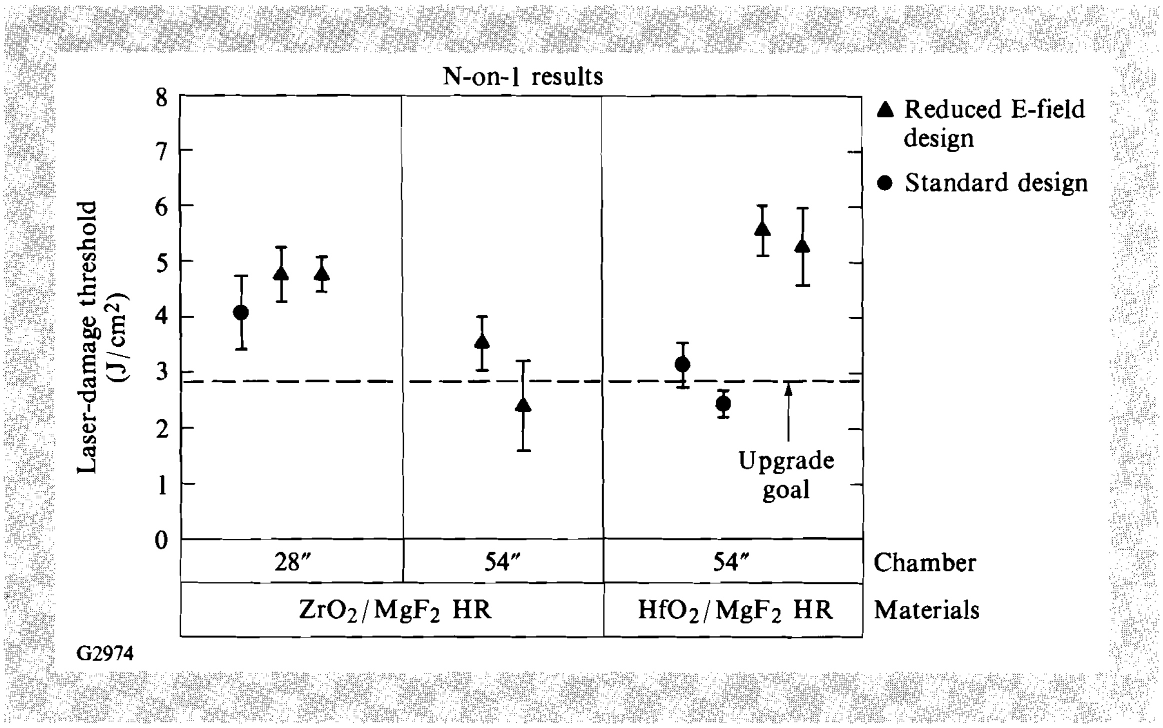

Fig. 44.36
Coatings produced with MgF_2 for the low-index layer show significant variation in LDT. Both ZrO_2/MgF_2 and HfO_2/MgF_2 high reflectors show promise for use at OMEGA Upgrade fluence levels.

Special emphasis was placed on the use of scandium oxide since a significant amount of promising damage data has been obtained with this material. However, the cost of the material is so high that its use would be prohibitively expensive for the OMEGA Upgrade. In an attempt to reduce the cost of scandia coatings but still retain the best qualities of the material, we tested several hybrid designs. These consisted of a multilayer next to the substrate consisting of HfO_2/SiO_2 (15 layers), followed by a 10-layer stack of a Sc_2O_3/SiO_2 multilayer. The damage-test results for a standard design and a reduced E-field design are seen in Fig. 44.37. Results for the same reactive-evaporation (RE) design, but with MgF_2 substituted for the silica, are also given. The thresholds are unimpressive for most of these coatings with few exceeding the upgrade requirement. The unusually small error bar implies a distinct onset of damage, which may indicate a damage mechanism different from that in two-material HR's. Physical properties such as stress, hardness, and adhesion between layers are all probable causes for coating failure. It is possible the multiple material combinations exacerbate these problems.

Most of the tested coatings meet OMEGA Upgrade optical and mechanical requirements. Some of the coatings made with magnesium fluoride are softer and scratch more easily, but survive well in a benign laboratory environment. Multilayers made with low-voltage ion assist are the hardest coatings. The most promising coatings are the hafnia/magnesium fluoride

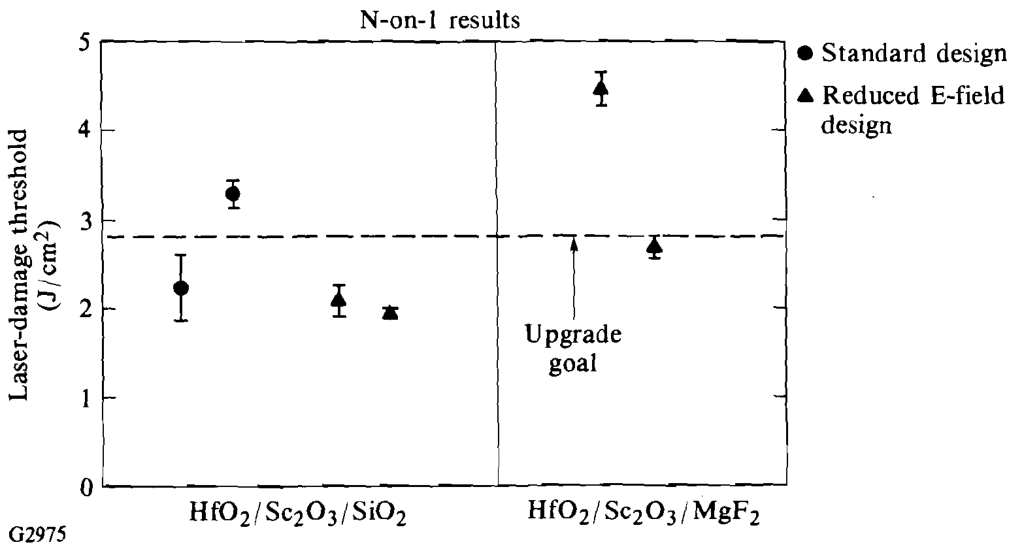

Fig. 44.37
The LDT of hybrid high reflectors using scandia as an upper-stack component. Although scandia generally provides a promising LDT in HR coatings, its performance in the hybrid-HR designs was disappointing.

and hafnia/silica coatings. Future work will gather data for non-normal incidence-angle coatings for both hafnia designs and some zirconia designs. More data will be gathered to document differences in thresholds due to equipment used and method of operation.

Ion Assist of High Reflectors

Ion-assisted deposition (IAD) is a combination of evaporation and ion beam-sputtering techniques. In a standard evaporation geometry (see Ref. 9) with an electron-beam source, ions from a broad-beam source[10] are directed toward the substrate. The ions with a kinetic energy about 100 times higher than the evaporant ions can effect chemical and physical changes in the films. Resultant coatings have significantly altered structures, reduced void fractions, less roughness, and less absorption. To investigate the effect of ion assist on the damage threshold of HR coatings we used low-energy (200-eV) oxygen ions. Structural changes to the film were kept to a minimum by maintaining low ion-beam currents.

Early tests with IAD coatings consistently gave poor damage results. Since average absorption in these films was measurably reduced, local contamination from the ion source was suspected of lowering the thresholds. The ion source consists of a discharge chamber with a tungsten filament, accelerator grids composed of graphite, and another tungsten filament for beam charge neutralization. To test for contamination, we ran the gun without using the accelerator grids (just the filaments on, producing negligible grid erosion), and then with two levels of accelerator-grid erosion. The results for a zirconia/silica HR made in the 28-in. chamber are given in Fig. 44.38(a). A slight improvement in the damage threshold is seen for both the negligible- and low-erosion cases, while a steep drop in threshold was observed when the grid erosion was high. A final coating was made with a different gun equipped with molybdenum grids with settings to give low erosion. This coating gave the highest threshold for this material pair seen in the 28-in. chamber.

The experiments with IAD were continued at this point on the larger, 54-in. chamber with the molybdenum grid ion source. This chamber is equipped with a planetary-substrate rotation causing intermittent ion bombardment at the substrate level. The peak ion-beam flux impinging on the substrate was similar to the continuous flux measured in the smaller, 28-in. chamber. Coatings with both negligible erosion and low-erosion IAD of ZrO_2/SiO_2 layers were found to have thresholds similar to conventionally deposited substrates [Fig. 44.38(b)]. A coating made with IAD of only the ZrO_2 layers had a slightly higher damage threshold than all other tested zirconia/silica coatings. These near-optimum IAD conditions were used to make the hafnia/silica reflectors already described in Fig. 44.35. While the superior damage performance of the hafnia coatings cannot be solely attributed to IAD, it is clear that no detrimental effects from IAD were observed.

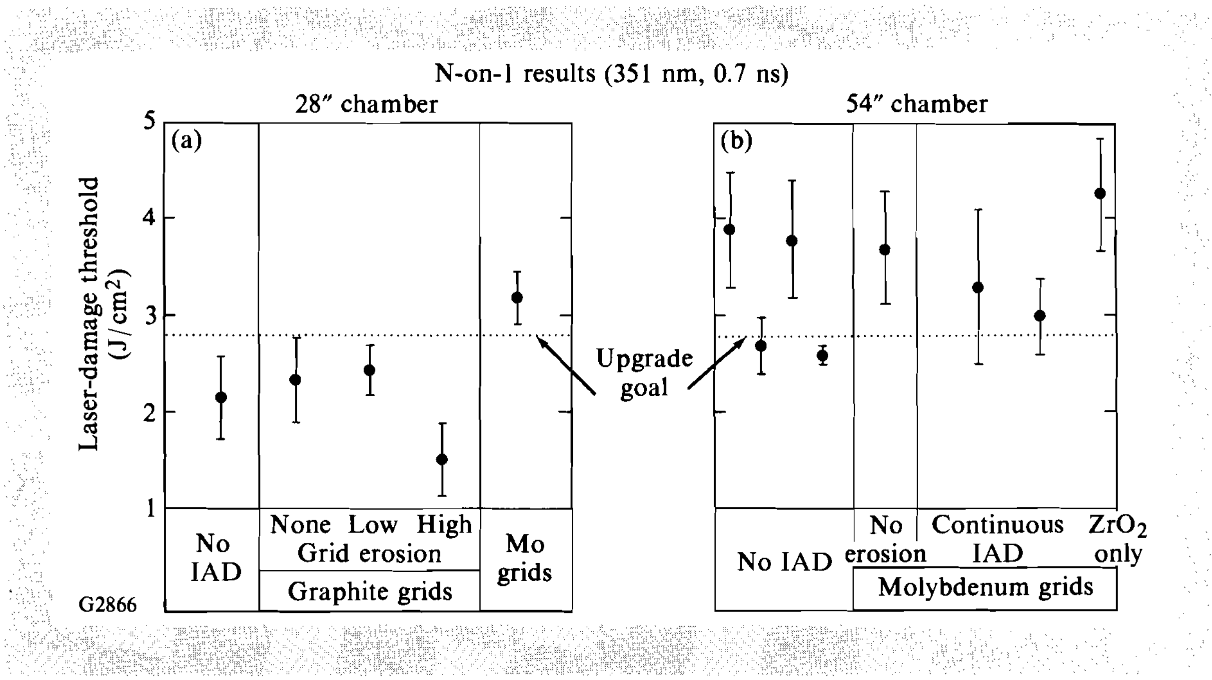

Fig. 44.38
LDT results for ZrO_2/SiO_2 coatings under various conditions of ion assist. Fig. 44.38(a) shows that contamination from graphite grids in the ion source was probably responsible for lower thresholds. Coatings made with molybdenum grids showed the highest threshold for these materials in the 28-in. chamber. Figure 44.38(b) shows little change for the ZrO_2/SiO_2 HR when the ion source is operated continuously. However, operating the source during only the high-index ZrO_2 layer results in a higher threshold.

The use of IAD adds a large number of parameters to the deposition process, including ion-source design, electrical settings, and gas choice. By making reasonable choices for these parameters, guided by past experience with IAD-modified films and laser damage, we have made better films in terms of both damage resistance and mechanical properties. These films were made in a production-sized coater under intermittent bombardment. While scale-up of all properties still must be demonstrated, we believe this will be a straightforward extension of current technology.

Sol-Gel Coatings

Early tests of dielectric coatings made with physical vapor-deposition methods for OMEGA Upgrade applications were disappointing (Fig. 44.39). Coatings using conventional, IAD, or reactive-ion plating (RIPD)[11] did not meet threshold goals for the upgrade. Sol-gel coatings[12] have been shown to meet both damage and optical requirements of laser systems similar to the OMEGA Upgrade. We tested several coatings supplied by Lawrence Livermore National Laboratory (LLNL) and Rutherford labs in England. All coatings had fairly good thresholds with two easily exceeding upgrade damage goals. A polymeric vapor-penetration barrier coat over one of the Rutherford samples (labeled COLSI/OC) drastically reduced the threshold.

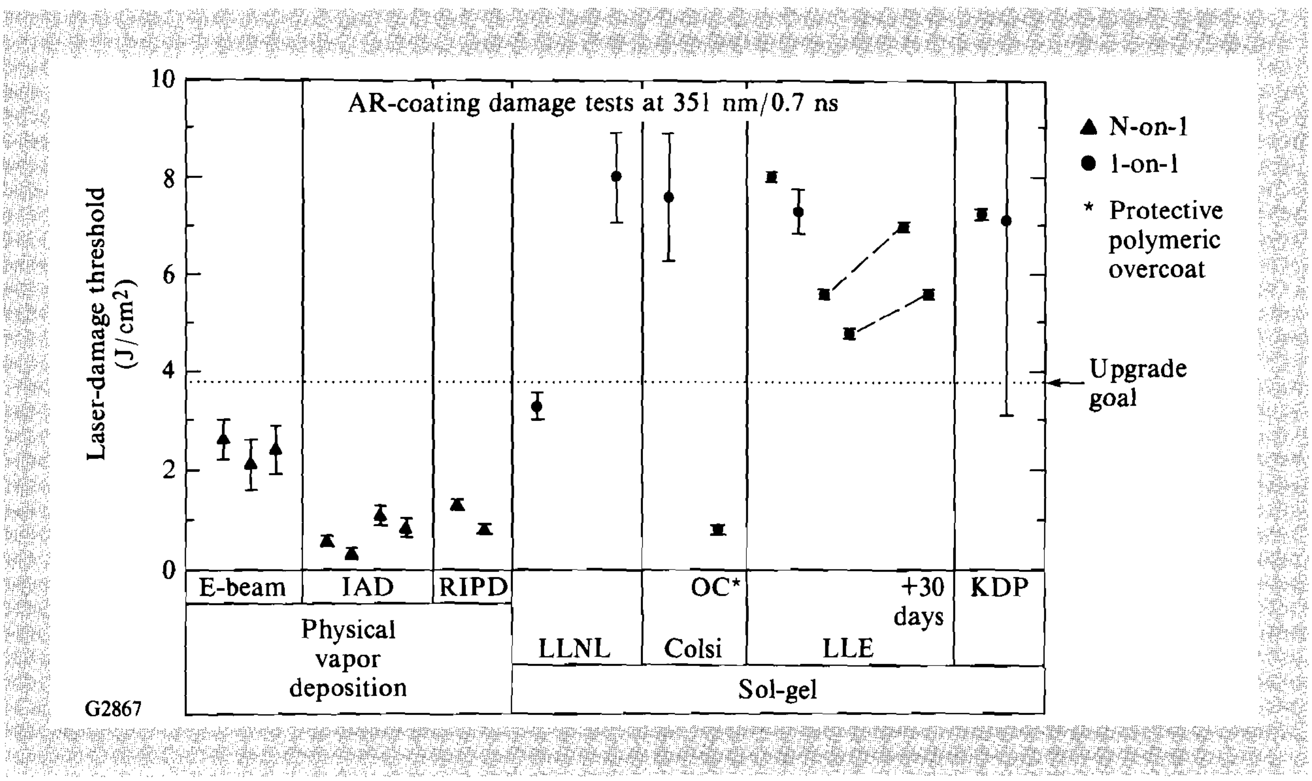

Fig. 44.39
LDT results of AR coating produced both by physical vapor-deposition techniques and sol-gel methods. Some of the sol-gel coatings were tested after a period in a laboratory environment. No degradation was observed in these coatings. All coatings were applied to silica substrates with the exception of the potassium di-hydrogen phosphate (KDP).

We developed a capacity to produce and purify the sol-gel solution, and spin a controlled thickness onto damage-test substrates. Coatings were applied to fused silica cleaned by the same aqueous method described for high reflectors. The coatings were spun in a class-100 clean room. Several pieces of potassium di-hydrogen phosphate (KDP) were also cleaned (using non-aqueous solvents) and sol-gel coated. The damage results for these samples (Fig. 44.39) show that most of these coatings tested well above OMEGA Upgrade requirements. Two samples were retested after standing exposed for 30 days in a typical laboratory environment. Neither coating exhibited deleterious effects after aging.

Sol-gel development has continued with the emphasis on scale-up problems. A dip-tank arrangement has been assembled and successfully tested with substrates up to 30 cm in size. Small prototype tooling has been designed and tested for two-sided spin coating of substrates. Also, work is proceeding on design of full-size spinner tooling. Prototypes of bonded KDP crystals for the OMEGA Upgrade will be coated. To date, the bonding material integral to the design of the frequency-conversion crystals (FCC) has been analyzed and may prove to be a contamination problem for the sol-gel coating. Further development is required in this area.

Distributed Phase Plates

The existing distributed phase plates (DPP's) for the OMEGA laser were made by evaporating the phase-retardation pixels onto a flat optic through a patterned photoresist coating.[13] These devices were adequate for current OMEGA peak fluences and had the advantage that the underlying antireflection and DPP coating could be removed. This process did not meet OMEGA Upgrade damage requirements as seen in Fig. 44.40. Damage tests were conducted on a patterned and unpatterned optic coated with an evaporated silica retardation layer. The antireflection coating was omitted to improve the damage characteristics of the device. All of these devices failed to exceed the damage criteria for the upgrade.

The new method for fabricating DPP's involves etching the pattern into the silica substrate. The threshold of these etched devices should be higher since there is no possibility of trapping contaminants within or underneath an evaporated layer. Unless the substrate surface is damaged during the etching process, a threshold comparable to that of an uncoated fused-silica surface will be attained. The patterns were etched into photoresist-patterned substrates using the same ion sources as described in the section on IAD. The working gas for etching was argon, which assured a longer filament life for the ion sources. The substrates were rotated during deposition, and the ion source was pointed off center, which reduces the etch rate but provides for better uniformity across the plate. After etching, the photoresist is removed with a solvent, and the surface is cleaned. Both a sol-gel AR-coated and an uncoated etched DPP were laser-damage tested. Test sites included both the "on" and the "off" hexagons in the array. The thresholds (Fig. 44.40) are among the highest we have tested for any surface at 351 nm, 0.7 ns. The tests were limited by the failure of the index-

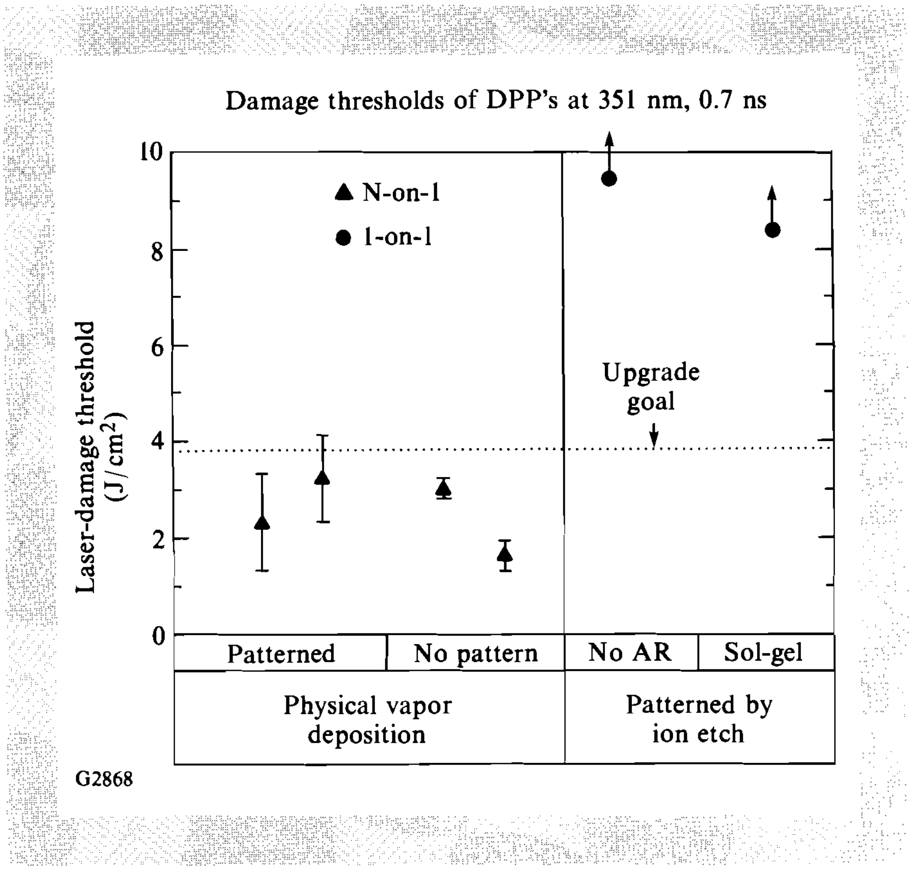

Fig. 44.40
The thresholds of the distributed phase plates (DPP's) (left) are well below the required level, even in the ideal case of a SiO_2 layer directly deposited onto a silica substrate without a pattern. Conversely, the thresholds of the etched DPP's approach those of polished uncoated surfaces.

matching fluid for the rear surface of the optic, so only a lower bound on the threshold is available. The pixel edge structure is also an improvement over the old DPP's. The magnified edges of both the etched and evaporated DPP may be seen in Fig. 44.41. The smoother, well-defined edges of the etched DPP will reduce scatter and also reduce the effect of size mismatch between the on and the off pixels. There was no effect on the phase difference when the sol-gel AR was applied to the DPP.

Future work on DPP's will focus on scale-up and etch-thickness control of the ion-etched DPP's. Up to now, damage-test pieces were created with a small, 3-cm ion source in a bell-jar vacuum system. Existing focused and defocused 8-cm ion sources will be characterized, modeled, and optimized for the best geometry for high uniformity across a clear aperture of 30 cm. The sources will be installed in existing chambers for uniformity testing and etch-rate measurement. A monitoring system will be developed for unattended automatic etching of the DPP. Photoresist coating and processing technology will also be developed for the upgrade apertures.

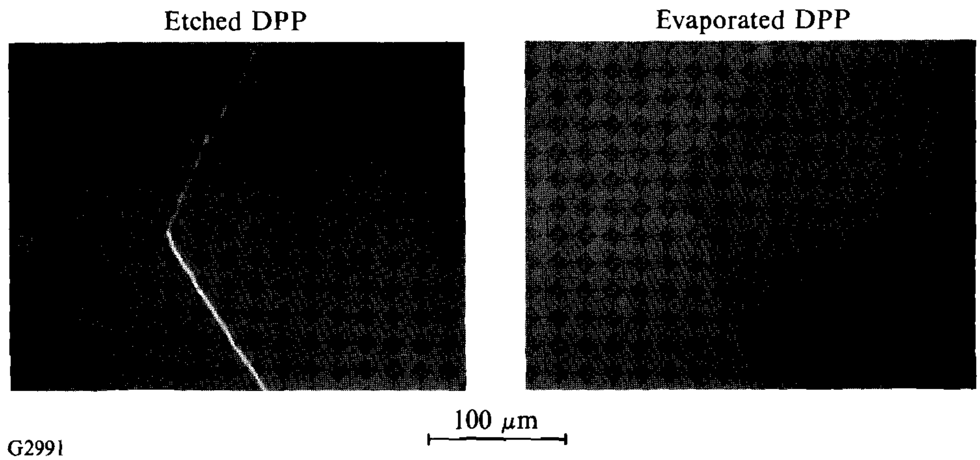

Fig. 44.41
Etched DPP's have better edge definition (left) than DPP's made with the deposition process. Higher edge definition produces less scatter, especially in smaller DPP patterns.

Conclusions and Further Study

Small-scale prototypes of all 351-nm components have been fabricated and have laser-damage thresholds exceeding the design requirements of the OMEGA Upgrade. Further testing and coating will provide better statistics for reflectors at both normal and high-incidence angles. Other considerations of scale-up such as scatter, stress, coating endurance, and damage properties of large aperture coatings will be explored. Development of sol-gel coating will continue with emphasis on the problems in coating the top-hat design of the KDP frequency-conversion crystals, large focusing aspheres, and blast windows and water jackets for the single-segmented disk amplifiers. Finally a significant effort will go into scaling the etched-DPP technology to OMEGA Upgrade-sized apertures.

ACKNOWLEDGMENT

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