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Laser Conditioning of Optical Thin Films*

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Results are presented that show the damage thresholds of e-beam deposited multi-layer $\text{HfO}_2/\text{SiO}_2$ thin films can be permanently increased by a factor of 2 to 3 by illumination with subthreshold fluences of laser light. This subthreshold illumination procedure is referred to as "laser conditioning". The films used in this study were prepared by three different physical-vapor-deposition techniques: ion-beam sputtering, plasma plating and e-beam evaporation. Only the e-beam deposited films showed consistent and significant improvement with laser conditioning. Of the material pairs examined ($\text{HfO}_2/\text{SiO}_2$, $\text{ZrO}_2/\text{SiO}_2$ and $\text{TiO}_2/\text{SiO}_2$), $\text{HfO}_2/\text{SiO}_2$ gave the greatest and most consistent damage improvement with conditioning. The number of layers and the reflective or transmissive characteristics of the $\text{HfO}_2/\text{SiO}_2$ films were found to have little impact on laser conditioning of the film. The results show that the damage thresholds of a wide range of e-beam deposited coatings (e.g. HR's, polarizers, etc.) can be improved by laser conditioning. Several possible conditioning mechanisms are examined.

Key words: "annealing", band-gap states, conditioning, damage, damage thresholds, dielectric oxides, laser conditioning, laser damage, optical thin films, paramagnetic states, point defects.

1. Introduction

At LLNL we are improving processes for fabricating damage resistant optical coatings for use on high-peak-power lasers such as those used in inertial confinement fusion (ICF) research. The coating fabrication processes under study are physical vapor deposition (PVD), plasma assisted chemical vapor deposition (PCVD), and sol-gel processing. Each alternative has certain advantages. However at the present time only PVD involving e-beam evaporation in vacuum is a fully scaled and practical process capable of fabricating the high-quality, large-aperture optical coatings needed for ICF research.

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A significant limitation on the operating fluence (or peak power) of high-peak-power lasers is the laser damage threshold of the optical components. Over the last two decades laser conditioning, or "annealing", using sub-threshold illumination has been reported as a method to increase the damage threshold of optical thin films and bulk optical materials [1-10]. In most cases, details of the process and materials involved have been sketchy. In addition, little effort has been made to apply such a process to improve the threshold of optical components installed in operating laser systems. In general, two mechanisms have been proposed as responsible for the observed conditioning effects. The first mechanism involves "laser cleaning", whereby volatile contaminants (especially atmospheric water) are removed either from the surface [1-4, 7, 8] or from the bulk of the material [5, 9]. It has been proposed that absorption by these contaminants leads to laser damage. This conditioning process appears to be reversible for surfaces [3] but permanent for bulk materials [9]. The second mechanism involves laser heating of the material until a change in crystallinity is observed [6] or polishing damage is removed [10]. The mechanism applicable to a particular system appears to be dependent on the optical material and the laser conditioning wavelength.

It has been shown by Wilder and Thomas that dielectric multilayer coatings similar to those used as turning mirrors in the LLNL Nova laser system show a laser conditioning effect [5]. In our study a number of different optical coatings were prepared in an attempt to identify the importance of different film parameters on both conditioned and unconditioned damage thresholds for materials used on the Nova laser. These film parameters are: dielectric material, deposition method and multi-layer film design. In the following pages we examine each of these three parameters. In addition we examine possible changes in the coating material that may lead to laser conditioning. Specifically we present evidence that absorption/desorption of contaminants and laser induced phase changes are not responsible for the conditioning effect observed in the films studied here. Also we observed that the conditioning effect is permanent for these films. Finally in a concluding section we propose an electronic defect mechanism to explain the laser conditioning effect in the dielectric films.

We intend to employ conditioning in the near future to increase the damage threshold of one meter diameter replacement mirrors for the Nova laser. Fabrication of these new mirrors with $\text{HfO}_2/\text{SiO}_2$ highly reflective (HR) coatings is scheduled to begin during the 1990 fiscal year.

2. Damage testing

The "figure-of-merit" for coating serviceability continues to be damage threshold. Damage thresholds measured at 1064 nm on three different facilities at LLNL are reported here. The repeatability on different lasers adds to the credibility of our conclusions. The Nd:YAG laser systems used are:

- i. A variable pulselength laser which operates in a single shot mode with pulselengths varying from 1 to 16 ns.
- ii. A rep-rated laser that operates at pulse-repetition frequencies (PRF) of 10, 15 or 30 Hz with a pulselength of 10 or 16 ns.

iii. A rep-rated laser that operates at 18 Hz with a pulselength of 8 ns and is capable of rastering large aperture optics automatically.

The laser fluence is determined using calorimetry measurements and the measured beam profile recorded with a CCD video camera. Commercial software is used to calculate the peak fluence from these data. The laser spot size is usually greater than 1 mm diameter ($1/e^2$). Damage is defined as any visual change in the sample after laser irradiation when viewed by 100X Nomarski microscopy. To observe the on-set of microscopic damage, photographs of the test area are taken before and after testing and compared for evidence of damage to the surface. A detailed description of damage testing apparatus and procedures is presented in an accompanying paper in this symposium by Rainer et al. [11].

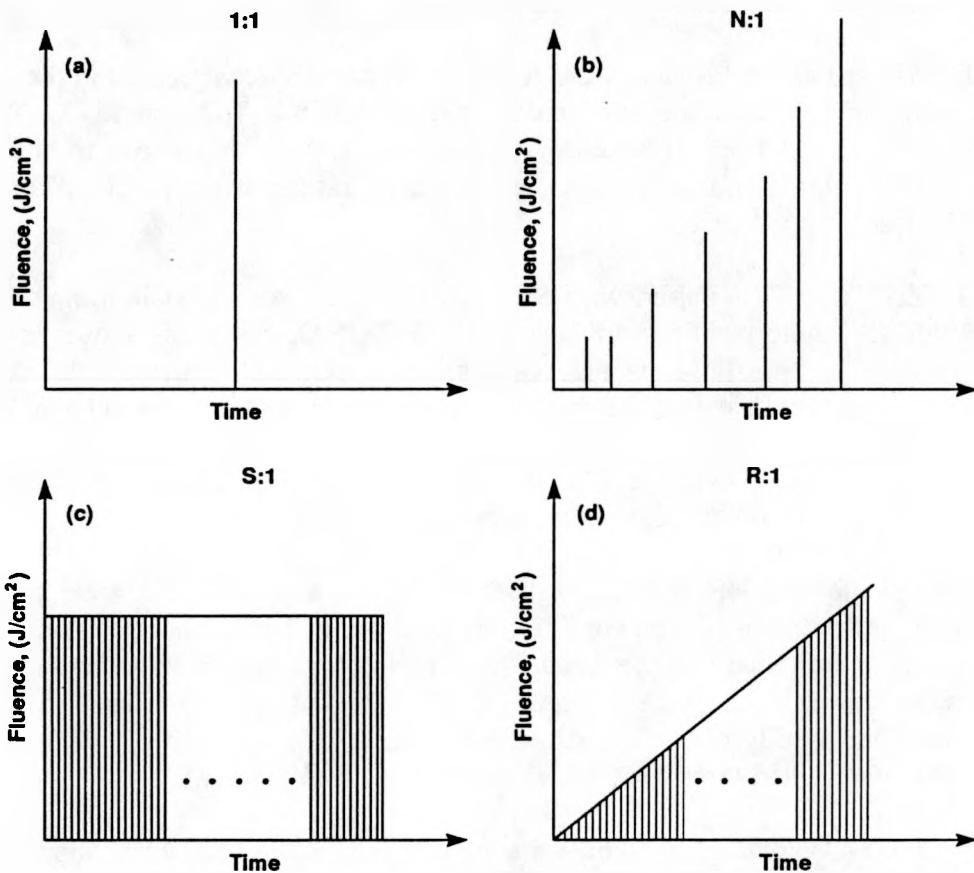


Figure 1. Four methods used to measure damage threshold: (a) single shot per site (1:1), (b) multiple shots per site with large increments in fluence between shots (N:1); (c) multiple shots per site at constant fluence (S:1), and (d) multiple shots per site with a ramped increase in fluence (R:1). Note that N:1 and R:1 tests give "conditioned" damage thresholds.

The damage thresholds reported here were determined by one or more of the four test sequences shown in figure 1. The four test sequences differ in the number of shots, the time between shots, and the range of fluences used. The damage threshold measured by 1:1 and S:1 shot programs are reported as "unconditioned" thresholds; these shot programs expose

each site to only one fluence level. The 1:1 test uses a single shot per site. The S:1 test uses a series of constant-fluence shots on each fresh (i.e. unexposed) site with the time between shots being short (equal to 1/PRF). Conditioned thresholds are measured by R:1 and N:1 tests. The R:1 test uses a series of shots separated by short intervals as in S:1, but the fluence is varied from zero to a pre-set upper bound in a linear ramp. The N:1 test uses a series of single shots, the fluence of each shot is increased step-wise until a pre-set upper bound is reached. The time between shots for N:1 testing can be rather long, generally several minutes. Uncertainty in the threshold measurement is typically $\pm 15\%$.

3. Results and discussion

3.1 Damage threshold dependence on film materials

SiO_2 was used as the low-index dielectric for all multilayers prepared in the course of this work. High-index dielectrics considered were: HfO_2 , ZrO_2 , TiO_2 and Ta_2O_5 . Ta_2O_5 was rejected because of the need to bake coated parts in O_2 after deposition to achieve full oxidation and low absorption. Baking at elevated temperatures is not practical for massive (250 kg) Nova-scale optics.

HfO_2 , ZrO_2 and TiO_2 paired with SiO_2 were directly compared in simple "quarter-wave" HR designs, the optical performance of the $\text{HfO}_2/\text{SiO}_2$ coating is shown in figure 2. Inconsistent damage thresholds were obtained from $\text{TiO}_2/\text{SiO}_2$ mirrors fabricated both during this study and in the past. The formation of highly absorbing sub-oxides of TiO_2 [12] is difficult to avoid during high vacuum deposition and may be the reason for the inconsistent damage thresholds. Therefore TiO_2 was also eliminated as a potential film material, leaving only HfO_2 and ZrO_2 as high-index materials of interest.

E-beam evaporated HR coatings of both $\text{HfO}_2/\text{SiO}_2$ and $\text{ZrO}_2/\text{SiO}_2$ were prepared in order to study the influence of laser conditioning on the damage threshold. Upon conditioning, both material combinations produced a marked improvement in damage threshold for 1-ns pulses, as shown by the data in figure 3. These results indicate that $\text{HfO}_2/\text{SiO}_2$ and $\text{ZrO}_2/\text{SiO}_2$ have essentially equivalent damage resistance; the unconditioned thresholds are 6-8 J/cm^2 and conditioned thresholds are as much as 3 times greater.

The $\text{HfO}_2/\text{SiO}_2$ coating thresholds are very reproducible and show about the same factor of improvement due to conditioning over a broad pulselength range, as shown in figure 4. The reproducibility of the threshold measurements was remarkably consistent considering three different laser-damage-test systems were used in the study. Based on the data in figure 4 for the $\text{HfO}_2/\text{SiO}_2$ coatings, the damage threshold scales with pulse-length as approximately:

$$\text{unconditioned: } D_t \simeq 7 t_p^{0.35}$$

$$\text{laser conditioned: } D_t \simeq 19 t_p^{0.30}$$

where D_t is the damage threshold in J/cm^2 and t_p is the laser pulselength in ns. This pulselength scaling of the damage threshold falls within the range typically reported by

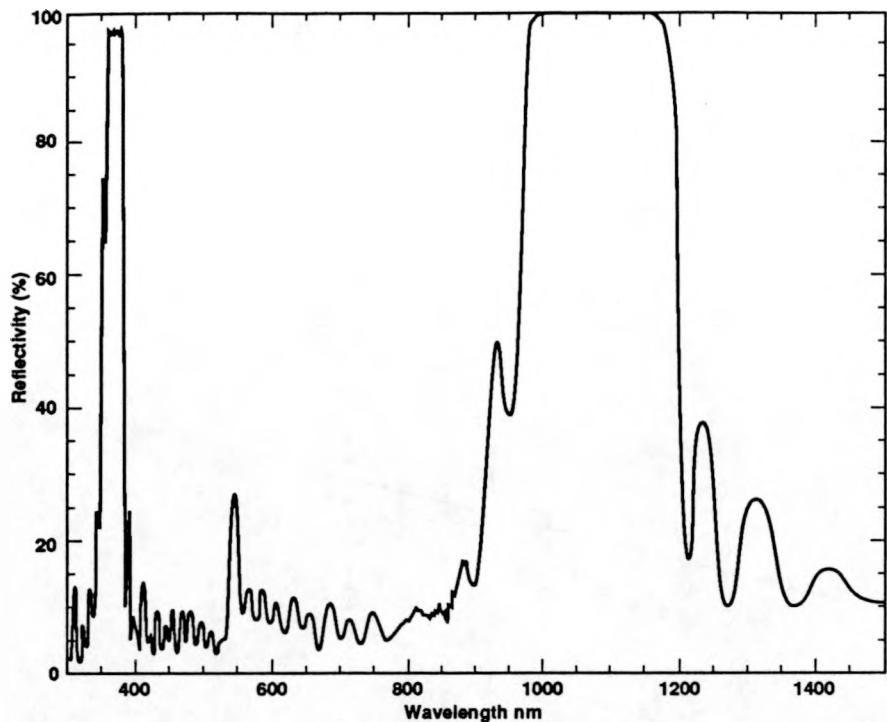


Figure 2. Measured reflectance vs. wavelength for a simple $\text{HfO}_2/\text{SiO}_2$ quarter-wave stack that was prepared as part of this study. The reflectance at 1064 nm is > 99.5%.

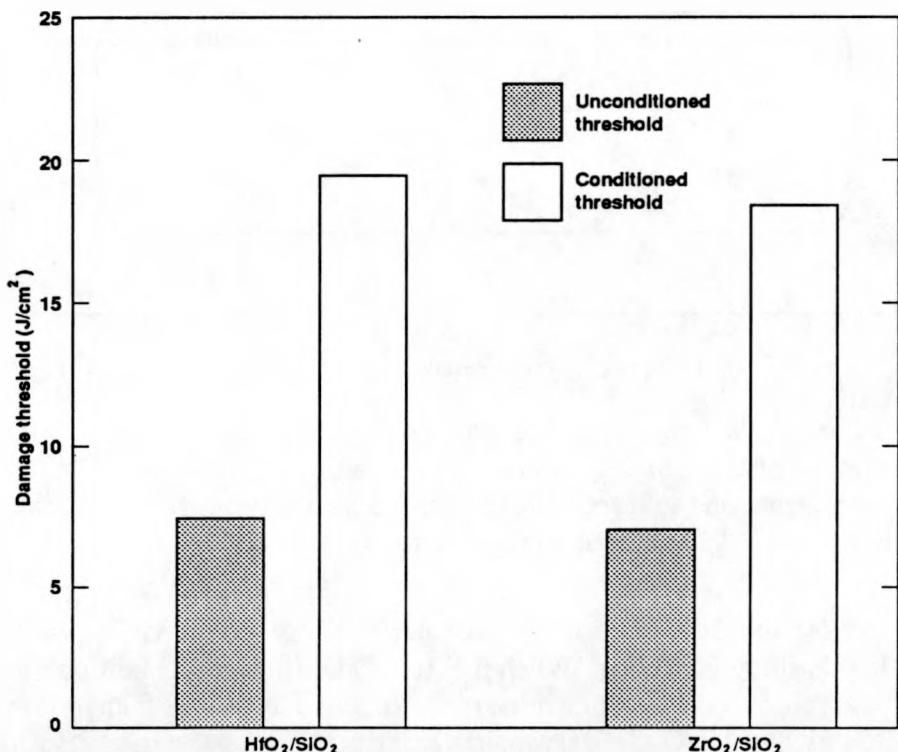


Figure 3. Comparison of unconditioned and laser-conditioned damage thresholds for $\text{HfO}_2/\text{SiO}_2$ and $\text{ZrO}_2/\text{SiO}_2$ HR quarterwave stacks for 1-ns pulses, 1064 nm. The unconditioned values are from 1:1 tests and the conditioned values are for N:1 measurements.

others studying damage to thin films (i.e. between about $t_p^{0.25}$ and $t_p^{0.4}$) [see for example reference 13].

In contrast to the $\text{HfO}_2/\text{SiO}_2$ thresholds, we found that the $\text{ZrO}_2/\text{SiO}_2$ coatings showed significant site-to-site variability in threshold measurements. In some tests the $\text{ZrO}_2/\text{SiO}_2$ showed marked improvement with conditioning (as in figure 3) while in other tests they showed no improvement.

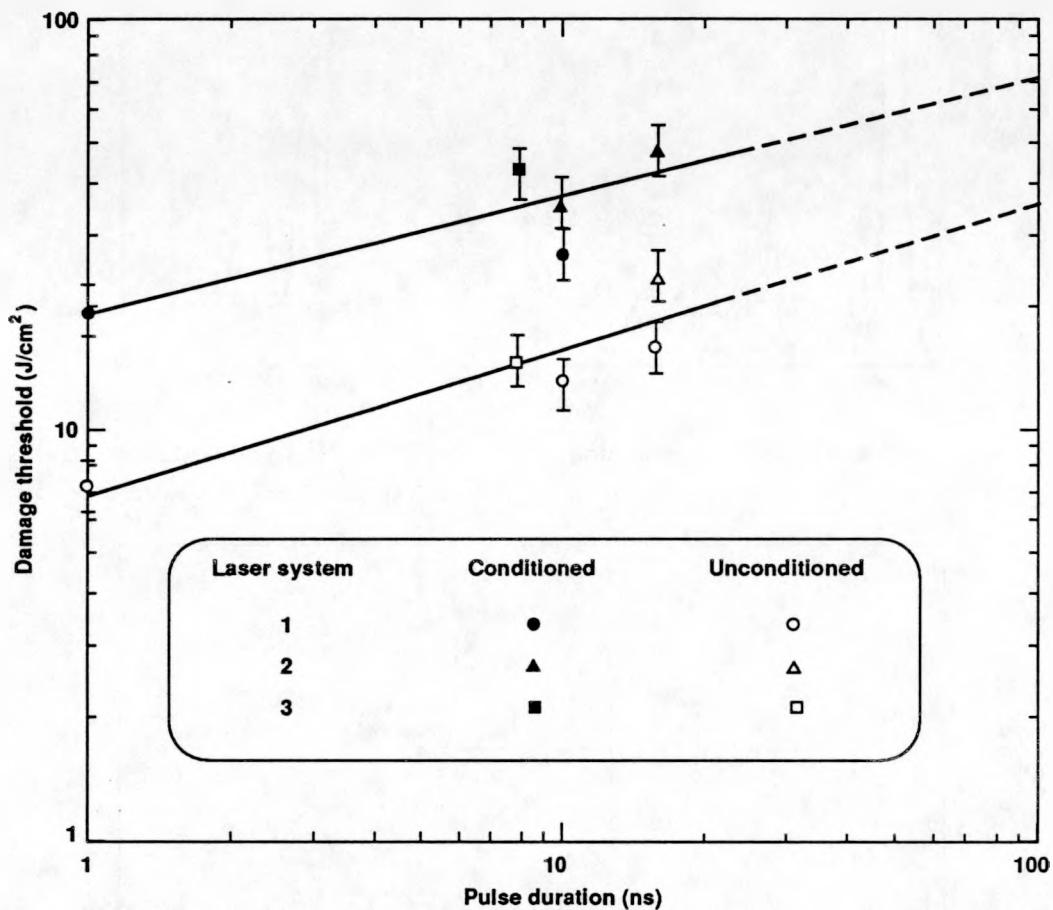


Figure 4. Measured pulselength scaling for conditioned and unconditioned damage thresholds of $\text{HfO}_2/\text{SiO}_2$ quarterwave HR coatings at 1064 nm. The data are from measurements on the three different laser systems described in section 2 and the solid line represents a least squares fit to the data.

The reason for the difference in damage performance of the ZrO_2 - vs. HfO_2 -based coatings used in this study is unclear. When the $\text{ZrO}_2/\text{SiO}_2$ films did condition, the threshold improvement was comparable to that observed for the $\text{HfO}_2/\text{SiO}_2$ films. We therefore suggest that $\text{HfO}_2/\text{SiO}_2$ and $\text{ZrO}_2/\text{SiO}_2$ have similar intrinsic film properties. However, in the case of ZrO_2 , the damage threshold is often controlled by extrinsic properties such as visible defects. These extrinsic defects lead to the site-to-site variability observed in the damage threshold. Using Nomarski microscopy, we found that the ZrO_2 - and HfO_2 -based films have about the same density of visible defects; however, the defects tend to be somewhat larger in

the $\text{ZrO}_2/\text{SiO}_2$ films. As was pointed out by Kozlowski et al. [14], it is unlikely that modest differences in size or density of defects, *per se*, leads to different damage threshold. Rather these differences probably indicate different types of defects that may be more or less susceptible to either laser damage or conditioning or both.

3.2 Damage threshold vs. film deposition method

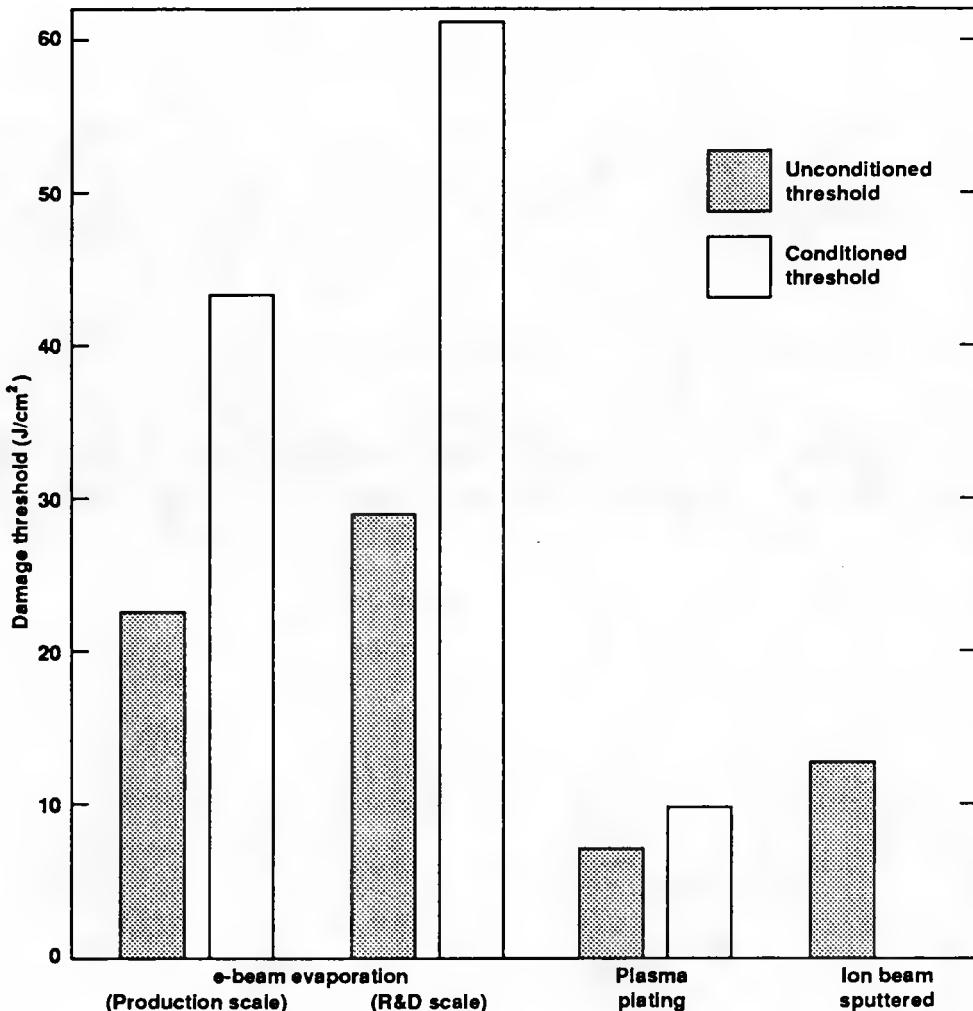


Figure 5. Unconditioned (S:1) and conditioned (R:1) damage thresholds (16 ns, 1064 nm) for $\text{HfO}_2/\text{SiO}_2$ HR coatings prepared by e-beam evaporation, plasma plating, and ion-beam sputtering. The e-beam data are for coatings prepared on both the small scale R & D coater and the large scale (3-m diameter) production coater.

During the course of this study we prepared HR films of $\text{HfO}_2/\text{SiO}_2$ by three different deposition techniques: (1) conventional e-beam evaporation, (2) ion-beam sputtering (IBS), and (3) plasma plating (PP). In addition, for the case of e-beam evaporation, we examined the effects of scale-up by preparing coatings in a small (1.2-m diameter) research-scale chamber as well as the large 3-m diameter production-scale chamber used to fabricate the current 1-m diameter, 250-kg Nova mirrors.

The choice of IBS and PP as coating methods was based on the observation that coatings produced by these methods have less porosity (i.e. higher density) and therefore have greater mechanical strength and environmental durability [15-19]. This latter quality was thought to be particularly important because of previous reports of film conditioning (and laser damage) being related to water-vapor absorption [1, 3, 4]. However, our measurements show that HR films of $\text{HfO}_2/\text{SiO}_2$ deposited by IBS and PP have lower damage thresholds than those made by conventional e-beam evaporation. Figure 5 compares the measured damage thresholds of coatings made by e-beam evaporation, IBS and PP. Laser conditioning does not significantly increase the damage threshold of the PP coatings. In addition, threshold measurements of IBS films were very inconsistent and the conditioning effect was not always observed, consequently no laser conditioned value is reported in figure 5. In this study laser conditioning was consistently observed only in e-beam-deposited coatings. In addition we note that the film absorption at 1064 nm, measured by calorimetry [20], was 500-5000 ppm for PP, 100-250 ppm for IBS and ~100 ppm for e-beam deposition.

Figure 5 also compares measured damage thresholds for coatings made in a small and a large e-beam coating chamber. The results show that conditioning is observed for the films produced in these two chambers; therefore the conditioning characteristics of the $\text{HfO}_2/\text{SiO}_2$ films are preserved in scale-up. The conditioned and unconditioned thresholds for the coatings produced in the small coater are 30-40 % higher than those made in the production coater, however.

3.3 Damage threshold dependence on coating design

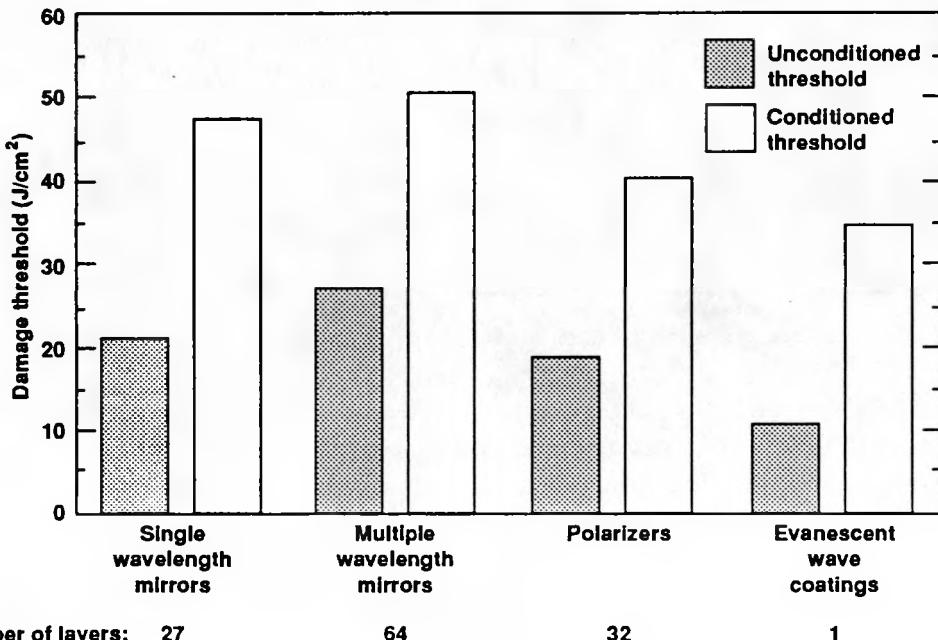


Figure 6. Comparison of conditioned and unconditioned damage thresholds for $\text{HfO}_2/\text{SiO}_2$ multilayer HR's, polarizer and single-layer evanescent wave coatings; all data are for 1064-nm laser irradiation. The HR data are for pulse lengths of 16 ns and the polarizer and evanescent wave coatings are for 10-ns pulses.

The data in the previous section describes laser conditioning effects in HR coatings consisting of a simple optical quarterwave stack of high- and low-index materials. We also found that the conditioning effect occurs in both transmissive and reflective coatings and in single as well as multilayer films. In other words, the conditioning effect appears independent of the coating design for $\text{HfO}_2/\text{SiO}_2$ film materials.

Figure 6 shows the unconditioned and conditioned damage thresholds for: (a) single-wavelength (1064 nm) HR coatings each consisting of a 27-layer quarterwave stack, (b) complex, 64-layer trichroic HR coatings designed to reflect at 1064, 532 and 355 nm, (c) 1064-nm polarizers with a rejection ratio of about 300 to 1, and (d) a single-layer SiO_2 evanescent wave coating 1400 nm thick. Note that for this series of coatings there appears to be no correlation between the threshold improvement via conditioning and the coating design. In addition, the degree of improvement by conditioning is also largely independent of the number of layers (or interfaces) within the coating for these $\text{HfO}_2/\text{SiO}_2$ coatings.

3.4 Effects of material changes on laser conditioning

Laser conditioning of thin films has previously been linked to the removal of adsorbed water or other atmospheric contamination [1, 3, 21-23]. When such a "cleaning" mechanism was reportedly involved, the enhancement of the damage threshold by conditioning was often found to be temporary [3].

Experiments performed at LLNL on $\text{HfO}_2/\text{SiO}_2$ mirrors indicate that the laser conditioning effect is permanent for these films and not related to the presence, or removal, of atmospheric water. To demonstrate this, $\text{HfO}_2/\text{SiO}_2$ mirrors made by conventional e-beam deposition were allowed to equilibrate with ambient air (20°C, relative humidity ~50%) and were then damage tested at 1064 nm. Both the unconditioned and conditioned damage thresholds were determined. The coatings were then desiccated for 10 weeks over P_2O_5 to remove physically absorbed water [24]. The removal of water was observed as a shift in reflectance to shorter wavelengths (table 1). The occurrence of spectral shifts in optical filters upon drying is well known [25]. The conditioned and unconditioned thresholds of the dried coatings were then measured and compared to the thresholds of the same samples before they were dried. No change in threshold was observed (fig. 7). Furthermore, the

Table 1. Measured Short Wavelength Shift of $\text{HfO}_2/\text{SiO}_2$ HR Coatings After Drying Over P_2O_5 for 10 weeks

<u>Deposition method</u>	<u>Spectral Shift (nm)</u>
E-beam evaporation: R&D-scale chamber	25
E-beam evaporation: Production-scale chamber	5
Ion beam sputtering	7
Plasma plating	3

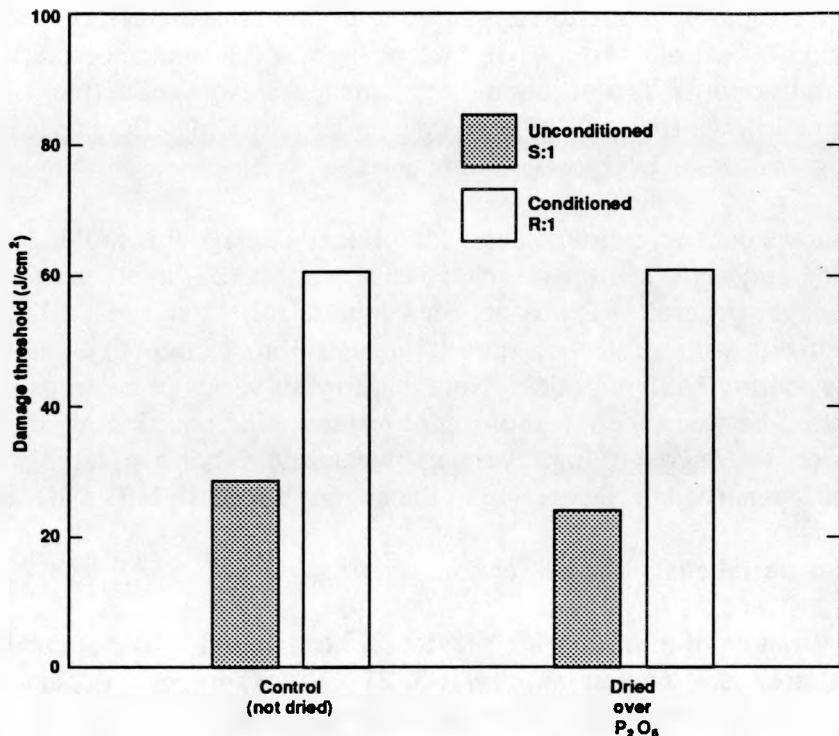


Figure 7. Conditioned (R:1) and unconditioned (S:1) damage thresholds (1064 nm, 16 ns) for $\text{HfO}_2/\text{SiO}_2$ HR coatings equilibrated in ambient air and desiccated over P_2O_5 .

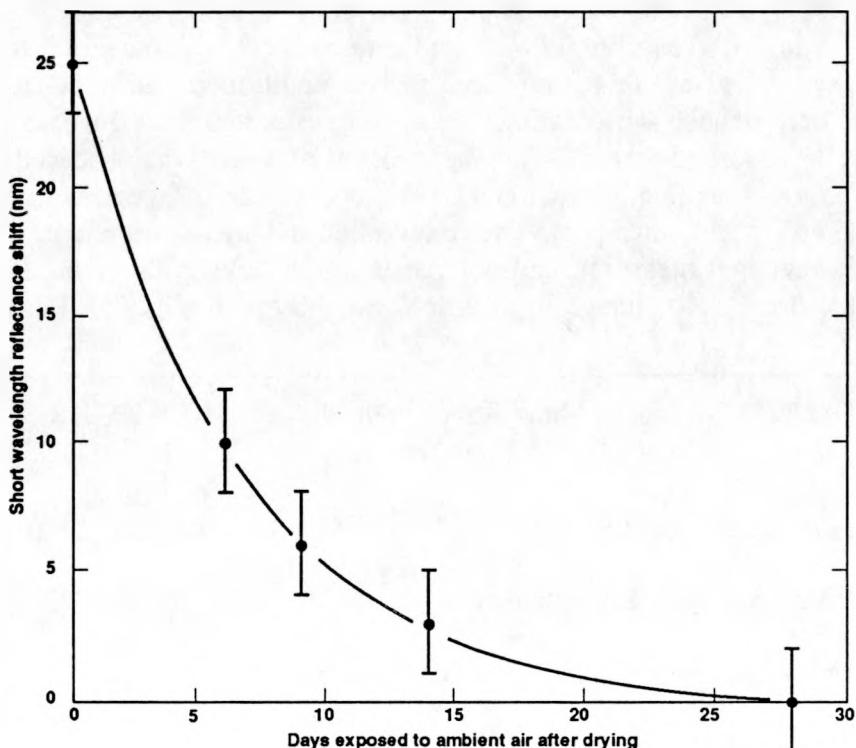


Figure 8. Time dependence of the spectral shift, to short wavelength, of a desiccated $\text{HfO}_2/\text{SiO}_2$ HR upon exposure to air.

reflectance shift caused by drying was found to be reversible. When the dried coatings were re-exposed to moist air their reflectance shifted back to longer wavelengths and the original performance was recovered. Figure 8 shows the gradual shift in the reflectance at 950 nm of one of the dried mirrors as it was re-equilibrated with ambient air. The optical absorption at 950 nm was used to monitor the slight shift in coating performance because a pronounced minimum in reflectance occurred at this wavelength (fig. 2). This structure allowed the slight reflectance shift to be accurately monitored. The time scale for equilibration is about 4 weeks. (Measurement of water absorption at 3350 cm^{-1} verified that the change in reflectivity was due to changes in the water content.)

The role of photolytic vaporization of contaminants in conditioning was investigated also. We attempted to "clean" and thus condition $\text{HfO}_2/\text{SiO}_2$ mirrors by exposure to broadband flashlamp light [26]. The lamp output fluence is about 10 J/cm^2 in 0.5-ms pulses, although it is over a very broad spectral range (~ 350 to 2000 nm). No increase in damage threshold was observed for the mirrors processed in this way, we therefore tentatively rule out "photolytic cleaning" of contaminants as a conditioning mechanism. Additional discussion of these experiments as they relate to electronic defects is given by Kozlowski et al. [14].

X-ray analyses of the $\text{HfO}_2/\text{SiO}_2$ HR coatings also indicate that conditioning does not involve any obvious material phase change. A diffractometer scan of an unconditioned mirror (using $\text{Cu, K}\alpha$ x-rays with wavelength of 1.5405 \AA) is shown in fig. 9a. Peak assignment indicates that the HfO_2 is partially crystalline. The crystalline fraction appears as mostly the monoclinic phase and a trace of cubic present on a high amorphous background. TEM analysis has shown that the HfO_2 layers are initially amorphous, but as the deposition continues, the crystal size increases so that at about halfway through the deposition columnar growth is observed. In contrast, SiO_2 is entirely amorphous. Diffractometer scans of the conditioned and unconditioned coatings showed no detectable change ($\pm 5\text{-}10\%$) in the quantities or types of phases present (fig. 9). Therefore we conclude that no phase change, that is measurable by this technique, accompanies laser conditioning of these coatings.

As a final note we report that the enhancement of damage threshold by laser conditioning is permanent in the $\text{HfO}_2/\text{SiO}_2$ e-beam deposited films. To determine this, a large area ($\sim 4\text{-cm}$ diameter) of an HR coating was conditioned using the Nova laser. The conditioned threshold of this sample was then monitored by subsequent small-aperture tests over a period of about 10 weeks. Details of this experiment and the results are given in the companion paper by Kozlowski et al. [14]. The results indicate that the preconditioned threshold was increased > 2 times over the unconditioned threshold and remained unchanged, within experimental error, over a period of 10 weeks. As shown above (fig. 8), the time scale for complete reabsorption of water in a desiccated film is only about 4 weeks. Thus film conditioning caused by laser induced water (or other contaminant) desorption, followed by reabsorption can be eliminated. We conclude that for all practical purposes the conditioning effect is permanent.

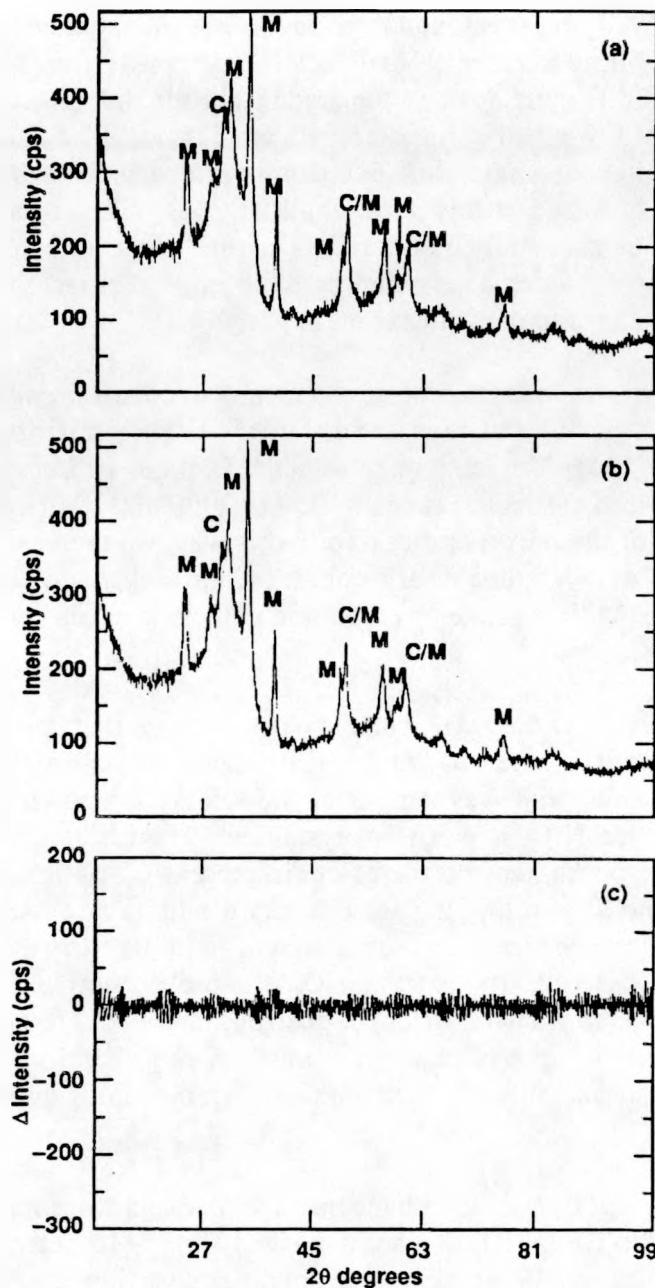


Figure 9 X-ray diffractometer signal vs. 2θ for (a) an unconditioned and (b) a conditioned $\text{HfO}_2/\text{SiO}_2$ HR coating. 'M' and 'C' identify peaks associated with the monoclinic and cubic phases of crystalline HfO_2 , respectively; (c) the difference between the two spectra as determined by subtracting (b) from (a).

3.5 Thoughts on the mechanism for laser conditioning

Before proceeding to describe what may cause the conditioning effect, it is worthwhile to state what it does not depend on (at least for the films discussed in this work). The

results in the preceding sections indicate the conditioning effect is:

- i. not associated with the removal of absorbed moisture or other atmospheric contamination,
- ii. not clearly dependent on the number of coating layers or the transmissive or reflective characteristics of the optical design,
- iii. not associated with measurable recrystallization within the film.

The results therefore indicate that the often proposed "laser cleaning" or phase change mechanisms are not the dominant mechanisms responsible for the conditioning phenomena occurring in the dielectric films of interest here. In a paper by Edwards et al. [27] we further show that linear absorption by contaminating particles [5] does not cause damage. We instead propose below that the laser conditioning effect observed in high quality optical thin films is associated with intrinsic electronic defects in the films.

Of the materials we have tested, conditioning is only consistently observed in $\text{HfO}_2/\text{SiO}_2$ films deposited by conventional e-beam evaporation. One significant characteristic of e-beam coatings, that distinguishes them from films made by other processes, is the low energy of the material being deposited ($\ll 1$ eV). In addition, the condensation process is one of extremely rapid quenching from a high temperature vapor to a much cooler substrate $\sim 100\text{-}300^\circ\text{C}$; this can be thought of as "splat cooling". This rapid quenching can result in porous films dominated by intrinsic defects [28]. We believe that the "non-equilibrium" state of these e-beam films is associated with their laser conditioning properties. In the following paragraphs we speculate on the nature of these intrinsic defects and on how they may be associated with the laser conditioning process.

We propose that the laser damage of unconditioned films deposited by e-beam evaporation occurs by a four step process:

- i. photo-excitation of electrons from shallow electronic gap states into the conduction band,
- ii. excitation of the free carriers to high energy by acceleration under the optical electric field or by free carrier absorption,
- iii. subsequent transfer of the excess energy to the lattice via avalanche [29] or an electron-phonon interaction (lattice heating) [30],
- iv. heating of the film to some critical damage temperature such as the melting or boiling point of the dielectric material.

The importance of conduction band electrons to optical damage is not a novel concept. In fact, the dependence of the laser damage threshold on the presence of conduction electrons has been demonstrated for bulk SiO_2 [30], as well as for silicon and glass [31]. The novel concept is the connection between the conduction band electrons and the condi-

tioning effect. We suggest that laser conditioning occurs due to the removal of the source of conduction band electrons in step (i) above. That is, when the dielectric material is illuminated at a low fluence, the electrons in the defect levels are excited to the conduction band. Since the optical electric field is not large enough to cause damage (steps (ii) to (iv)) the electrons decay into deep levels from which they cannot be easily excited into the conduction band on subsequent illuminations. When the 'conditioned' dielectric is subsequently illuminated at intensities above the unconditioned damage threshold, the number of electrons available for transfer to the conduction band is low and, therefore, the net energy transferred to the lattice is too low to cause damage; hence the observed laser conditioning effect.

E-beam deposited dielectric coatings are known to have a high concentration of electronic defects in the band-gap [32]. Point defects in amorphous and crystalline SiO_2 are well characterized because of their importance to solid state electronic technology [33-35]. Very little is known however about similar defects in other wide band gap dielectrics such as HfO_2 , ZrO_2 , and TiO_2 . As shown schematically in figure 10, energy bands in amorphous solids can be thought of as distorted by local variations in bonding. In general, this produces discrete states in the band gap that are distributed randomly in the amorphous material [36]. Wide band gap dielectrics deposited by e-beam evaporation are highly disordered and electronically metastable. A high concentration of occupied gap states located above the Fermi level may result from this type of disorder. An example of such defects, paramagnetic point defects, have been observed in amorphous SiO_2 [37]. It has been demonstrated that these states can be filled by electrons as a result of UV illumination and then subsequently emptied by thermal treatment [38].

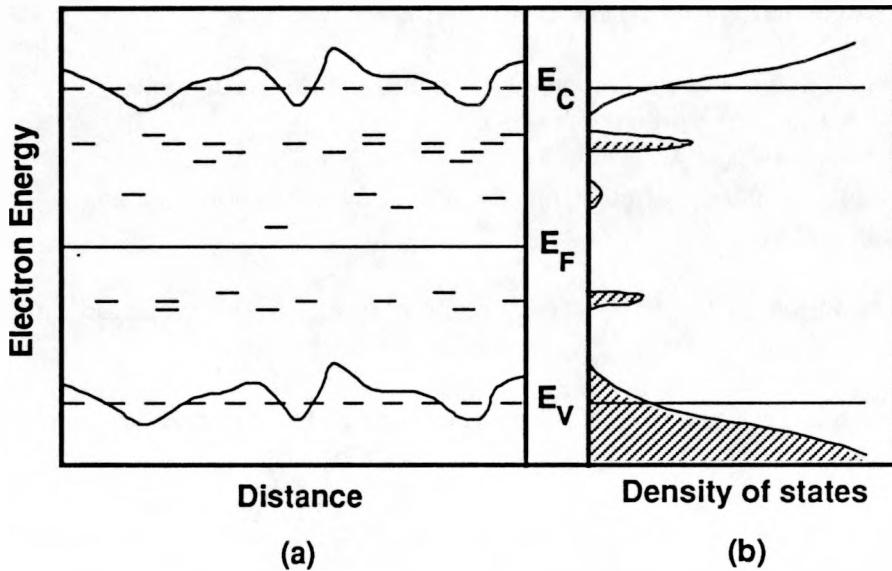


Figure 10. (a) Schematic diagram showing the model representation of the energy bands of an amorphous dielectric solid including gap states. E_v , E_f and E_c refer to the energies associated with the valence band, Fermi level and conduction band, respectively; (b) schematic representation of the corresponding density of states.

The independence of the laser conditioning effect on film design and number of layers suggests that the electronic defects of interest are bulk defects and not associated with interfaces. Further discussion of the nature of the electronic defects and the mechanism by which energy is transferred to the crystal lattice is given by Kozlowski et al. [14].

As shown above, "non-equilibrated" films produced by e-beam deposition produced consistent conditioning effects. In contrast, films of SiO_2 prepared by high temperature PCVD [39, 40] have very high unconditioned thresholds, comparable to those for conditioned films of e-beam deposited SiO_2 . We believe that the PCVD process produces inherently more equilibrated films, which are not susceptible to the damage/conditioning mechanisms important to unconditioned e-beam coatings. Unfortunately the PCVD coating technology is not yet developed to the scale required to fabricate large-aperture optical components for fusion research lasers.

Because of their non-equilibrium deposition conditions, the plasma plating and ion-beam sputtering processes examined in this study might be expected to produce electronically highly defective films with a high concentration of electronic defects similar to those produced by e-beam deposition. The IBS and PP films, however, showed negligible improvement in the damage threshold upon laser conditioning. The low unconditioned threshold and lack of a conditioning effect suggests that a damage mechanism other than the electronic defect mechanism described above might be dominant for these films.

4. Conclusions

The results of experiments reported here show that optical thin films of SiO_2 or $\text{HfO}_2/\text{SiO}_2$ deposited by conventional e-beam processes can be laser conditioned to significantly increase their laser damage threshold. This is accomplished by illuminating the coating at fluences below the damage fluence. Our results indicate that:

- i. the damage threshold can be increased by factors as great as 2-3x,
- ii. the conditioning effect is permanent,
- iii. the conditioning effect is dependent on the dielectric material and the deposition technique, but shows little dependence on coating design or the size of the deposition chamber
- iv. conditioning may be related to the removal of electrons from intrinsic defects energetically close to the conduction bands in these materials.

Through the use of the conditioning process the damage threshold of e-beam deposited optical coatings can be made to exceed the operating fluences required by current and future fusion laser systems. Currently, e-beam deposition is the only process by which large-aperture, high-quality, optical coatings can be made. Results presented here show that the conditioning properties of e-beam deposited films are not influenced by scale-up to a production scale deposition chamber. In a paper by Kozlowski et al. [14] we further show that the laser conditioning process can be used to increase the damage threshold over entire

large area optical components. These laser conditioned e-beam coatings will comprise the next generation of large aperture optical thin films used on the LLNL Nova laser system.

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