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PREPRINTDamage Measurements on Optical Materials
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Damage Measurements on Optical Materials for Use in High-Peak-Power Lasers*

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Recently, we have expanded our efforts to develop state-of-the-art optical components for use in large-scale, high-peak-power, solid state lasers. Laser-induced damage to many of these components sets critical constraints on construction costs and limits the peak powers attainable in current and proposed devices. Increasing the damage threshold whenever possible by improving materials fabrication technology is therefore the ultimate goal of our work. The optical components and technological developments we are investigating are:

Highly reflective mirrors	• Kilo-layer coatings fabricated by high-temperature, plasma chemical vapor deposition (PCVD) • Laser conditioned physical-vapor-deposition (PVD) coatings • Multi-layer sol-gel coatings
Anti-reflective coatings	• Single layer sol-gel coatings deposited on substrates and non-linear crystals
Non-linear crystals	• High purity KDP, LAP and related frequency conversion crystals
Substrates and laser hosts	• High threshold surface preparation techniques • High threshold bulk materials
Polarizers	• Advanced PVD multi-layer coatings

Our research has resulted in an extensive database of laser-damage measurements. We present summaries of these measurements with particular emphasis on those taken at 1064 nm and 355 nm.

Key words: antireflective (AR) coatings; bare substrates; bulk damage; conditioning; damage; frequency conversion crystals; highly reflective (HR) coat-

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ings; laser-induced damage; plasma chemical vapor deposition (PCVD); polarizers; reflectors; sol-gel coatings; thin films.

1. Introduction

The damage-test capabilities at the Lawrence Livermore National Laboratory (LLNL) support and work in conjunction with a wide variety of laser programs. We conduct and contract out to the commercial sector research and development efforts to produce optical components with high damage thresholds for use in both high-peak-power and high-average-power laser systems. The high-peak-power work is in support of LLNL's Inertial Confinement Fusion (ICF) program which involves (1) the existing Nova laser system, (2) smaller ICF development laboratories, and (3) the Advanced Driver program for developing a successor to Nova. Much of this work has also overlapped with research conducted for the development of high-average-power laser systems including (1) the Medium Power Solid State Laser (MPSSL), (2) the free electron laser (FEL) program, and (3) the Laser Isotope Separation (LIS) program.

2. Laser damage-test capabilities

During the past year we have operated six dedicated damage-test facilities at LLNL, conducted full-aperture damage tests with the Nova laser, and also supported damage testing capabilities at several commercial locations. [1] Most of our lasers are single-shot or repeated Nd:YAG lasers operating at the fundamental wavelength, 1064 nm (1ω), or at one or more of its harmonics: 532 nm (2ω), 355 nm (3ω), and 266 nm (4ω). We also utilize excimer lasers for UV testing at 350 nm (XeF) and 248 nm (KrF). We operate with pulse durations ranging from 1 to 100 ns and at pulse-repetition frequencies (PRF) ranging from single shots up to 6000 Hz depending on the particular laser. Typically we conduct our tests with test-spot diameters of 1 to 2 mm, although we may resort to small spot measurements for very high fluences or also irradiate full-sized optics with one shot on LLNL's Nova laser. We conduct tests at a given site with one or more shots at a constant fluence or with successive shots at gradually increasing fluence (laser conditioning). This conditioning can raise damage thresholds of optical materials by factors of two to three. [2, 3]

3. Damage database

We have maintained and are expanding an extensive database of damage test results which cover 15 years of work at LLNL. [4, 5] In figure 1 we show a summary of these results for the past two years which represent 1300 tests conducted at 1064 nm with 1- to 16-ns pulses at PRF's ranging up to 120 Hz. The major portion of our efforts have concentrated on:

- (1) antireflective (AR) coatings fabricated by the sol-gel process;
- (2) highly reflective (HR) coatings fabricated by physical vapor deposition (PVD), plasma assisted chemical vapor deposition (PCVD), and the sol-gel process;
- (3) polarizers fabricated by PVD;
- (4) frequency conversion crystals; and
- (5) laser host and substrate materials.

During the past year we have re-established testing capabilities at the harmonics of Nd:YAG and are resuming testing of many of these same types of samples at these wavelengths also.

Figure 1. We have conducted over 1300 damage measurements in the past two years, most at 1064 nm with 1- to 16-ns pulses at PRF's ranging from single shots to 120 Hz.

Component:	Laser glass	Mirror	Polarizer	Frequency Converter	Focus lens	Debris shield
Coating:	None	HR	Polarizer	AR	AR	AR
Frequency:	1 ω	1 ω	1 ω	1, 2, 3 ω	3 ω	3 ω
Damage threshold specification at 10 ns (J/cm ²):	35	35	35	35 - 1 ω 25 - 3 ω	25	~25

Figure 2. A schematic diagram of a typical beam line for a large-aperture laser system shows the representative components. We are designing a system with components which must survive fluences of 35 J/cm² at 1064 nm (1 ω) and 25 J/cm² at 355 nm (3 ω) for 10-ns pulses.

4. High-peak-power laser requirements

We show in figure 2 the schematic of a typical beam transport system for a proposed large-aperture, high-peak-power, solid state laser system for future ICF research. For 10-ns pulses the various components in such a beam line must be able to survive fluences of 35 J/cm² if they are subject to 1 ω irradiation (laser glass, HR coatings, polarizers, frequency conversion crystals) and 25 J/cm² if they are subject to 3 ω irradiation (frequency conversion crystals, focusing lenses, debris shields). We show in figures 3 and 4 an overview of the current ranges of damage thresholds that we have achieved for these general categories at 1 ω and 3 ω respectively. These figures also provide information about specific materials, fabrication processes and irradiation conditions that allow us to achieve the desired design goals at each wavelength.

5. Specific threshold achievements

5.1 Highly reflective (HR) coatings

A significant portion of our efforts has concentrated on developing high damage threshold HR coatings by a variety of techniques (conditioned PVD coatings, kilo-layer PCVD coatings, and multi-layer sol-gel coatings). We have in fact achieved design goal thresholds with each technique as shown in figure 5. However, except for PVD coatings, much of our work is in the research stage and has not yet been scaled up to full-sized optics with dimensions ranging from 0.3 to 1 m. The damage thresholds of PVD HR coatings have gradually risen to levels at which they just barely meet our design goals. For several years numerous papers have reported on the improvement of laser damage thresholds of optical coatings by laser conditioning or "annealing" of the coatings. These past results have, however, reported only temporary improvement of damage thresholds to HR coatings. Recently we have found that HfO₂/SiO₂ HR coatings have yielded permanent threshold increases by factors of two to three to levels exceeding 50 J/cm² for 10-ns pulses and which in turn do not degrade with time. [3] Moreover we have shown that this improvement in threshold can be extended to full-sized optics. [2] The summaries shown in figure 5 show both the conditioned and unconditioned thresholds measured at 1064 nm for all of our highest threshold samples. A few tests were conducted with 1- or 16-ns pulse durations. We have determined that these coatings scale nominally as $\tau^{0.35}$ and have scaled the results accordingly to 10-ns values. [3] In some instances the damage thresholds were greater than the maximum fluence used for a particular test so that we only established a lower bound for these samples. These are indicated by arrows.

A promising technique we are investigating is the fabrication of PCVD HR coatings. [6] In this process alternate layers of slightly doped SiO₂ are deposited from the vapor phase plasma. The difference in index of refraction between alternating layers is relatively small requiring 1000 or more layers to produce an effective HR coating. Research samples produced by this process as well as a modified CVD process have yielded damage thresholds exceeding 60 J/cm² for 16-ns pulses at 1064 nm. We have plotted these data in figure 5 after scaling them to 10-ns thresholds. Most of the PCVD HR coatings were deposited on the inside of tubes nominally 1 cm in diameter. Threshold measurements were often limited by the poor optical quality of the drawn tubes rather than the actual HR coatings which are

Figure 3. Our damage threshold goal for a large-aperture, high-peak-power, fusion laser system is 35 J/cm² at 1064 nm (1 ω) for 10-ns pulses (grey vertical bar). The horizontal bars show our measured range of damage thresholds for various laser materials; laser "conditioning" of many of these materials places them near or above the threshold goal.

Figure 4. Our damage threshold goal for a large-aperture laser system is 25 J/cm² at 355 nm (3 ω) for 10-ns pulses as indicated by the grey vertical bar. The horizontal bars show our measured range of damage thresholds for various laser materials at 355 nm with 10-ns pulses. As in figure 3, conditioning significantly (and permanently) improves the damage thresholds of some materials.

approaching thresholds close to that of fused silica substrates. We are presently completing the construction of a coating chamber which will allow us to fabricate PCVD HR coatings on flat fused silica substrates up to 6" in diameter. [7]

Figure 5. Over the past two years we have developed higher threshold HR coatings at 1064 nm by using the three coating methods given above. The CVD and solgel techniques are still in a research stage (i.e. small aperture); PVD coatings have been fabricated for full 1-m applications. The data show the number of coating samples tested that gave the damage thresholds shown on the abscissa. Most tests were conducted with 10-ns pulses but some 1- and 16-ns tests were scaled to 10-ns thresholds by $\tau^{0.35}$.

We have successfully employed the sol-gel process for producing single layer AR coatings for many years and have scaled their applicability up to optics exceeding 1 m in diameter. We have extended this technology to produce multi-layer $\text{AlO}(\text{OH})/\text{SiO}_2$ HR coatings on 2"-diameter substrates. In the near future we will apply this technology to 8"-diameter substrates. We have measured damage thresholds of these coatings that exceed 40 J/cm^2 for 16-ns pulses at 1064 nm. To conform with the other data in figure 5 we have scaled these results to 10-ns values.

5.2 Antireflective (AR) coatings

Virtually all of the work that we have engaged in recently in the development of AR coatings has revolved around the deposition of single layers of SiO_2 by the sol-gel process

on fused silica, KDP, and other substrates. In figure 6 we show the results of tests conducted during the past two years at 1ω and 3ω . The tests represent a mixture of primarily 10- and 16-ns tests scaled to 10-ns values by $\tau^{0.35}$. Almost all samples we have tested in 1989 have comparable or higher damage thresholds than the best sol-gel AR coatings tested in the previous year. Moreover, in many of the recent tests we determined only lower bounds to the thresholds because we were either not able to achieve higher laser fluences for damage testing or because of massive damage to the blemished, uncoated rear surfaces of the substrates. It should be noted that most of the recent tests with thresholds $< 35 \text{ J/cm}^2$ were for research coatings which do not in fact represent the state-of-the-art of production technology. Routinely any sol-gel AR coating we fabricate at LLNL with known technology for high thresholds do in fact have thresholds exceeding 45 J/cm^2 . Only a limited number of tests were conducted so far at 355 nm with all exceeding the design goal of 25 J/cm^2 for 10-ns pulses. We were unable to damage two of the samples at fluences exceeding 38 J/cm^2 .

Figure 6. Damage thresholds of recent sol-gel AR coatings routinely exceed design goals for both 1064- and 355-nm tests. Earlier tests were conducted with 16-ns pulses which were scaled to 10-ns thresholds by $\tau^{0.35}$; more recent tests were conducted with 10-ns pulses.

5.3 Frequency conversion crystals

The conversion of laser beams from 1064 nm to shorter wavelengths is crucial for ICF applications. Frequency conversion crystals have always presented a significant problem from a damage standpoint in the construction of large-aperture laser systems. The crystals must first be grown to relatively large dimensions and yet be inclusion free to prevent bulk damage. The surfaces must then be figured and provided with AR coatings which are environmentally sound and yet keep losses and damage to a minimum. The latter problems have been routinely addressed with diamond turning and sol-gel AR coatings which can be

applied directly to the crystal surfaces at low temperatures. We have addressed the bulk damage issue by adhering to higher standards of cleanliness and the use of ultra-filtration of the constituent solutions used in growing conversion crystals. [8] We have conducted numerous damage tests on both deuterated and undeuterated versions of ℓ -arginine phosphate (LAP) and potassium dihydrogen phosphate (KDP). In figure 7 we show the results of damage tests on crystals which have been laser-conditioned using relatively large test-spot sizes (≥ 0.8 mm diameter). We have now standardized the technique to grow clean KDP crystals which have virtually no observable bulk inclusions and we are working on extending it to the growth of LAP. For both types of crystals we have measured 10-ns bulk damage thresholds exceeding 60 J/cm^2 at 1064 nm. At 355 nm these crystals meet or exceed the design goal of 25 J/cm^2 for 10-ns pulses.

Figure 7. Both LAP and KDP crystals can be grown to yield bulk damage thresholds $\geq 60 \text{ J/cm}^2$ at 1064 nm with 10-ns pulses. The samples meet or exceed the design goal of 35 J/cm^2 at 1064 nm and 25 J/cm^2 at 355 nm.

5.4 Bulk materials and bare polished surfaces

Most elements in the typical beam line of a large-aperture laser systems have some type of optical coatings deposited on them (AR, HR or polarizer). Essentially the only exceptions are the actual laser glass disks, mounted at Brewster's angle, and occasional lenses, beam splitters or windows where reflections or losses are not critical or where fluences exceed those that coatings can tolerate. At LLNL bulk damage issues for laser glasses have been addressed by an active campaign to reduce the inclusion of damage-causing platinum. [9] Other than frequency conversion crystals, the only other bulk materials typically included in the beam line are fused silica substrates or Faraday rotator materials. With the absence of inclusions, bulk damage to all these materials comfortably exceed 50 J/cm^2 and measurements often are indeterminate because of damage to surfaces at lower fluences. Recently we have conducted only isolated tests on bare surface samples.

[4, 5] In figure 8 we show a compilation of the average thresholds for a variety of common substrate materials with nominal indices of refraction near 1.5. This shows that bare surface thresholds scale as $\tau^{0.4}$ and are independent of material type. Since these are average values, they are in fact relatively conservative because a wide variety of substrate types and polishing techniques are included. Many of our typical fused silica measurements are from rear surface tests conducted on bowl-feed polished (super-polished) substrates. These results are ancillary to the primary intent of measuring thresholds of good quality AR coatings which had been deposited on the front surfaces of these same substrates. Rear-surface damage to bare surfaces is usually easier to observe than front-surface damage and sets a conservative limit on the sample since the effective fluences at the rear are higher than at the front. [12]

Figure 8. Average damage thresholds for many bare polished substrates with indices of refraction near 1.5 are independent of glass type and scale as $\tau^{0.4}$. The data are from references [4, 10, 11].

5.5 Polarizers

Polarizers consisting of multi-layer stacks of dielectric materials are typically fabricated with more complex coating designs using more layers than those used for comparable HR coatings. Unlike with an HR coating, the coating-substrate interface of a polarizer sees the full intensity of the transmitted laser beam. By their very nature, polarizers will also have greater angular sensitivity to each polarized component of the laser beam. These characteristics have usually combined to yield among the lowest thresholds for multi-layer coatings. We have recently conducted conditioning tests on a variety of $\text{HfO}_2/\text{SiO}_2$ polarizers from several vendors. In figure 9 we show that, as with HR coatings, we can expect to

find a significant improvement in thresholds by implementing laser-conditioning. From a limited database of conditioned polarizer tests we have observed an increase in threshold of about a factor of two to fluences $> 40 \text{ J/cm}^2$.

Figure 9. Laser conditioning of commercial PVD-fabricated polarizers improves their damage thresholds by an average factor of two to fluences $> 40 \text{ J/cm}^2$ for 10-ns pulses at 1064 nm.

6 Conclusions

We have constructed and utilized damage-test facilities at LLNL with which we have measured the damage thresholds for all crucial optical components necessary for the design of a multi-megajoule, high-peak-power, solid state laser system. Our results show that these materials can meet the damage specifications of 35 J/cm^2 at 1ω and 25 J/cm^2 at 3ω with 10-ns pulses. We have verified that the laser conditioning of multi-layer PVD HR's and polarizers can permanently raise 1064-nm damage thresholds above design goals and be scaled up to full-sized optics. Research-sized HR's can be fabricated by both PCVD and the sol-gel processes to exceed these goals and work is under way at LLNL to allow us to scale up this technology. Full-scale and research versions of sol-gel AR coatings on KDP crystals and fused silica substrates have reliably been operated and tested at fluences far in excess of design goals at both 1ω and 3ω . Careful attention to clean growth conditions has yielded KDP crystals with bulk thresholds comparable to those of fused silica. Bare and sol-gel-AR coated surfaces of fused silica and KDP have comparable thresholds at their respective harmonics and also exceed the laser design goals at both harmonics.

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