2/6-43 W.B.

(1)

MASTER

UCRL- 88156
PREPRINT
Conf-821170--3

UCRL--88156

DE83 012660

Raising the Surface-Damage Threshold of Neutral-Solution Processed BK-7 by Pulsed-Laser Irradiation

J. E. Swain
W. H. Lowdermilk
D. Milam

This paper was prepared for submittal to
Boulder Damage Symposium
Boulder, Colorado
November 15-17, 1982

Lawrence Livermore National Laboratory

Raising the Surface Damage Threshold of Neutral Solution Processed BK-7 by Pulse-Laser Irradiation*

J. E. Swain, W. H. Lowdermilk and D. Milam

Lawrence Livermore National Laboratory
Livermore, CA 94550

Gradient-index antireflecting surfaces can be produced by neutral solution processing of bare polished surfaces on borosilicate glasses. These processed surfaces have a median surface damage threshold of 12 J/cm^2 for 1-ns, 1064-nm pulses. The surface damage thresholds can be increased to fluences as large as 25 J/cm^2 by irradiating the surface with 1064-nm wavelength pulses at fluences below the initial threshold of 12 J/cm^2. For surfaces in their initial state, or surfaces subjected to repetitive laser irradiation, surface damage thresholds increased as the square root of pulse duration over the range of pulse durations from 1-20 ns.

Key words: neutral solution processing, laser damage, laser conditioning, antireflection films.

Schott Optical Glass Inc. recently developed a process for producing gradient-index anti-reflecting (AR) surfaces on borosilicate glass by leaching the glass in a nearly neutral solution of Na_2HAsO_4 in water [1]. This "neutral solution process" (NSP) was used to produce AR surfaces on BK-7 borosilicate crown glass, which is widely used for optical components of visible and near-infrared lasers. These NSP AR surfaces exhibited a median damage threshold of 12 J/cm^2 for 1-ns, 1064-nm pulses, which is more than twice the 5 J/cm^2 median threshold of silica-titania multilayer AR coatings [2]. Figure 1 compares these data. Recently we found that the damage threshold of NSP AR surfaces could be increased by as much as a factor of two by irradiating the surface with laser pulses at fluence levels slightly below the initial 12 J/cm^2 threshold.

Our experimental procedure for measuring laser damage thresholds has been described [3]. The procedure consists of irradiating a site on the surface of a sample with a pulsed Nd-glass laser beam approximately 2.5 mm in diameter. We examine the irradiated site, both before and after the laser shot, using a Nomarski microscope with magnification of 100. The sample is moved after each shot so that each site is irradiated only once. The fluence of the laser pulse is increased with each shot until damage occurs. The occurrence of damage is identified by a permanent change in the surface morphology observed using the Nomarski microscope. This measurement technique, in which each site on the surface is irradiated only once, is commonly called a "1-on-1" measurement.

The occurrence of damage in 1-on-1 tests of NSP AR surfaces was always accompanied by laser-induced emission of light. This light is probably recombination radiation from the plasma generated at the surface by the intense laser irradiation. Light emission from NSP AR surfaces was also frequently observed in the 1-on-1 tests at fluence levels slightly below those required to produce damage visible by microscopic examination. If, rather than moving the sample after each shot, we continued to irradiate the same surface area at fluence levels below the 1-on-1 threshold, light emission ceased after a small number of shots. The required number of shots was typically 1-8 and depended on the laser pulse duration. We found that after such treatment, the laser fluence could be raised to a level well above the 1-on-1 threshold before light emission or damage occurred. The threshold for damage determined by this procedure is called the "n-on-1" damage threshold.

We have measured 1-on-1 and n-on-1 thresholds for NSP AR surfaces using 1064-nm pulses at five pulse durations: 1,3,6,9 and 20 ns. Thresholds for a particular sample are shown in figure 2. Thresholds, either n-on-1 or 1-on-1 were defined to be the average of the least fluence that caused damage and the greatest fluence that caused no damage. The uncertainty depended on the separation of the two fluences which defined threshold, and on the uncertainty in fluence measurements themselves, $\pm$ 5%. In 1-on-1 measurements, front and rear surface damage thresholds were the same, as expected for a substrate with AR films on both surfaces. In the n-on-1 measurements with 6,9 and 20-ns pulses, rear-surface thresholds were less than front-surface thresholds, an observation for which we currently have no satifactory explanation. The

*Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under contract W-7405-ENG-48.

two straight lines drawn through the data have slope one-half. Therefore, over the range of pulse durations from 1 to 20 ns, thresholds of NSP surfaces scale in proportion to the square-root of pulse duration.

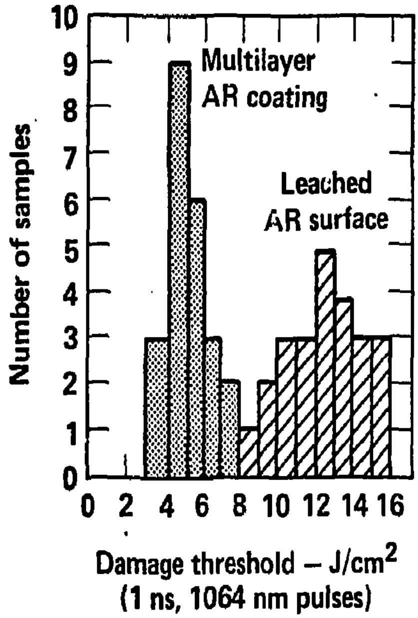

Figure 1. The median one-on-one surface damage threshold of neutral solution processed BK-7 is a factor of two higher than that of electron beam deposited dielectric AR coatings.

A possible mechanism for the increase in damage threshold measured in the n-on-1 tests is the removal of absorbing impurities from the surface by subthreshold irradiation. We believe absorption of energy from the laser pulse by particulate impurities is a common cause of damage to optical surfaces of transparent dielectric materials irradiated by laser pulses with near-ir or shorter wavelengths [4]. When irradiated initially at high fluence, as in the 1-on-1 tests, these particulates absorb laser energy and can reach temperatures above 1000°K, which is sufficient to induce surface damage by stress fracture or melting. When irradiated at lower fluence, the particles reach lower temperatures, which may be sufficient to promote desorption, but insufficient to produce damage. In some photographs of NSP AR surfaces taken with a scanning electron microscope, particles are visible in areas that were not subjected to n-on-1 irradiation, but absent from irradiated areas.

Another possible mechanism for removing particulates by low fluence irradiation is the emission of electrons or ions from the surface [5]. The quantity of charge emitted may be sufficient to substantially alter the static electric forces which normally bind particulate impurities to the surface.

The heating and desorption mechanism is, however, more consistent with our observation that "laser cleaning" is more easily accomplished using pulses of longer duration. When surfaces were irradiated with 20-ns pulses at subthreshold fluence, light emission usually ceased after one shot. Using pulses of shorter duration, more shots were required to "clean" the surface sufficiently that light emission ceased. At the shortest pulse duration used (1-ns), 6-8 shots were usually required. The maximum fluence at which the "cleaning" can be performed is that just below the 1-on-1 damage threshold, which, as shown in figure 1, increases as the square root of the pulse duration. Consequently, these results indicate that the total fluence accumulated over all the shots required to "clean" the surface and affect an increase in threshold is roughly constant.

In conclusion, we have found that the damage threshold of gradient-index AR surfaces of neutral-solution-processed BK-7 glass can be increased by as much as a factor of two by pulsed laser irradiation at fluence levels just below the 1-on-1 damage threshold. Damage thresholds of these AR surfaces increase with the square root of the laser pulse for pulse durations in the range from 1 to 20 ns, and the improvement in threshold due to "laser cleaning" occurs over this entire range of pulse duration.

We are grateful for technical assistance by S. E. Peluso and for assistance by Treva Janssen and Diane Kelly in preparing the manuscript.

References

[1] Cook, L. M.; Mader, K. H.; and Schnabel, R. "Integral antireflecting surfaces on silicate glass", U.S. Patent Application, Ser. No. 309,149, October 6, 1982.
[2] Cook, L. M.; Lowdermilk, W. H.; Milam, D. and Swain, J. E.; Applied Optics 21, 1482 (1982).
[3] Milam, D. SPIE, Vol. 140, 52-61 (1978).
[4] Lowdermilk, W. H. and Milam, D. IEEE J. Quant. Elect. QE-17, 1881 (1981).
[5] Logothetis, E. M. and Hartman, P. L. Phys. Rev. 187, 460 (1969).

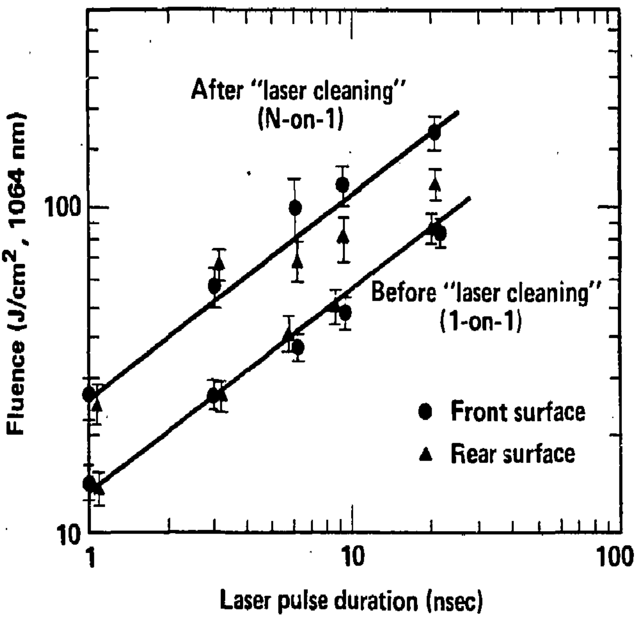

Figure 2. Pulse duration dependence of damage thresholds measured with 1064-nm laser pulses on the front surface and rear surface of a window with NSP AR surfaces. The straight lines are square-root functions. "Laser cleaning" produced n-on-1 thresholds that were nearly twice as large as 1-on-1 thresholds.