How to Polish Fused Silica to Obtain the Surface Damage Threshold Equals to the Bulk Damage Threshold

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Abstract

We compared the 1064 nm surface damage thresholds of fused silica polished by three different techniques:

1. A conventional polishing technique: that uses loose Alumina abrasives (lapping) followed by a fine Cerium oxide polish.
2. An alumina polishing process producing surfaces very close to super polished.
3. Process 2 followed by a silica polish until the silica surfaces are super polished.

We employed the same measurement technique that proved successful for the bulk damage threshold measurement to measure the damage thresholds of bare silica surfaces polished by the above three polishing techniques. We used an 8-nanosecond, single transverse and longitudinal mode pulsed laser, from a Q-switched Nd:YAG laser. We used the surface third harmonic generation technique to precisely place the focus of the laser beam on the surface of the fused silica window, and to measure the laser focus spot size which was found to be 8 μm in radius. Key findings include:

1. The surface damage threshold can be made equal to the bulk damage threshold. There is a large difference in single-pulse damage thresholds between bare silica

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surfaces polished using ceria, alumina only, and the surfaces polished using alumina followed by silica. The ceria polished samples have a statistical damage threshold ranging from 50 to 450 GW/cm^2., where as the alumina polished surfaces show damage at 200-500 GW/cm^2, with half the spots showing damage at the bulk threshold of 500 GW/cm^2. The windows polished by alumina followed by silica show damage almost universally at the bulk damage threshold of 500 GW/cm^2.

2. There are strong conditioning effects for these surfaces. The ceria polished surfaces have reduced thresholds for multiple pulses. The alumina polished surfaces attain the bulk damage threshold at most locations using multiple pulse annealing.
3. We found there was no beam size variation of the damage threshold irradiance for the bare alumina/silica polished samples.
4. By measuring the bulk damage threshold powers at different depth from the front surface, we have found experimental evidence of a self-focusing effect, which gives the correction factor for the measured bulk damage threshold.
5. We showed that surface irradiance is limited by breakdown of the silica and not breakdown of the air.
6. Damage morphologies were recorded for the different surfaces.

I. Introduction

Laser induced breakdown leading to optical damage in optically transparent material, such as fused silica, is important in the development of optics for high power laser applications. Earlier studies indicate that the polishing process of the substrate strongly affects the damage threshold of the surface [1]. However, this paper reported on polishing methods that were supposed to increase the damage threshold, but it did not give full information and cannot be relied on when designing optics for high power lasers. We believe better measurements of the optical breakdown threshold along with a better understanding of the optical polishing process are needed.

In our studies of optical breakdown on silica surfaces or in the bulk of silica, we attempted to answer a number of questions and also address several fundamental measurement issues:

1.Is the optical breakdown threshold in the nanosecond regime deterministic or statistical?

2.Is the surface optical breakdown threshold equal to that in the bulk?

3.What is the effect of the polishing process on the surface damage threshold of the bare surface?

4.Is the breakdown threshold irradiance independent on the size of the focus spot?

5. Experimental evidence of the self focusing correction factor of the bulk damage threshold.

6.Is the multiple shot damage threshold higher than the single shot damage threshold?

This paper is organized as follows:

Part II: Describes the polishing methods of fused silica.

Part III: Describes the property of optical breakdown

Part IV: Gives a description of our experimental set-up.

Part V: Discusses the temporal and spatial quality of the laser pulses.

Part VI: Describes the method of how to find the location and how to measure the focal spot size.

Part VII: Shows the surface damage thresholds.

Part VIII: Shows the damage morphologies.

Part IX: Discusses our conclusions.

II. Polishing methods of fused silica

The widely used method for polishing fused silica uses cerium oxide as a polishing compound. This polishing method leaves a thin film of ceria-silica on the surface of fused silica, to avoid this film a new method was adapted which uses alumina and silica as polishing compounds. These methods will be described below.

IIa. Cerium oxide polishing method

Cerium oxide is a typical medium used for polishing most optical materials; it is done following a loose abrasive lapping process using Al_2O_3. Cerium oxide slurries of

different sizes (from bigger to smaller) were used to polish the silica windows consecutively. Usually, the slurry of larger particle size is selected for the initial polishing, larger particles have more edges therefore have a higher removal rate. Once planarization has been achieved, the material removal rate slows considerably; it is important that all initial surface damage is removed with this step, prior to moving on to the next step. The majority of time spent in polishing is spent during the final polishing operation. In most cases, the larger-particle-sized slurry does not always result in the finish required for the final surface. Typically, a smaller-particle-sized slurry is selected for the final finishing step. The surface quality of the cerium oxide polished fused silica windows which were used in this test passed the 10/5 scratch/ditch (S/D) laser quality inspection.

IIb. Alumina polishing method

Alpine Research Optics (ARO) employed a loose alumina abrasive lapping process as a typical material removal processing, with the goal toward minimizing subsurface damage through-out the lapping/polishing processes. The lapping process was performed using a 9μ Al_2O_3 particles followed by 5μ Al_2O_3. After final lapping, the work surfaces were then evaluated with a 10x objective to be sure all macro damage was removed after each lapping step and that the surfaces would be clean enough to begin the pre-polish process. ARO chose to do the "Pre-Polish" step using Ultra Sol-M5-PS (an alumina polishing compound), and a 0.8-μ alumina slurry. The pH of the slurry was maintained at ~ 8 to prevent "Orange Peel"; which is the effect the acidic slurry can have on fused silica. After the "Pre-Polish" was completed, the fused silica windows were polished further with smaller sized slurries, the final step used a 100-nm slurry. The surfaces of the fused silica windows were evaluated to see whether they passed the 10-5 "Laser Quality" inspection. It was found that after the last alumina polishing run, the surface quality of the fused silica window was slightly better than the 10/5 S/D.

IIc. Alumina-Silica polishing method

After the silica window passed the 10-5 "Laser Quality" inspection, we began the final silica polishing process. For the final polish, we used the same polishing lap surface, a formulation of Gugolz pitch (optical quality polishing pitch) mixtures, submersed the

work in Ultra-Sol 500S (colloidal Si polishing slurry), a 78nm colloidal Si slurry. The Ultra-Sol 500 S was diluted such that 1 part Ultra-Sol to 50 parts of de-ionized water. The fused silica windows were polished until the desired goal of a very smooth surface was achieved; a target of less than 1Å roughness represents the smoothness of the polished surface. This target was selected so that we could be sure to achieve a level of finish on the surface that would closely represent the properties of the bulk material. To verify surface roughness, we used a phase shift interferometer [ADE Micro XAM], which indicated a 0.8Å surface smoothness profile.

III. Detection of optical breakdown

Intense laser light excites electrons from the valence band to the conduction band by three processes: tunneling ionization, multiphoton ionization, and impact ionization. When the plasma frequency of the free electron gas in the conduction band equals the laser frequency ω; then the density of the free electron gas is equal to the critical density n_{crit},

$$\omega_p^2 = \frac{e^2 n_{crit}}{m^* \varepsilon \varepsilon_0} = \omega^2 , \qquad (1)$$

where e is the electron charge, m^* is the effective electron mass, $\sum$ is the relative permittivity of the medium, and $\sum_0$ is the vacuum permittivity. For 1064 nm light, the critical density of fused silica is 2.08 x $10^{21}/cm^3$. At the onset of optical breakdown the following processes occur [1]:

a. A high-density plasma appears at the focus, and this plasma emits a white light (broadband). We detect this white light as the primary indicator of optical breakdown.

b. The dense plasma in the focal region absorbs, reflects, and scatters the incident laser pulse, causing a drastic drop in the transmitted laser power.

c. The HeNe probe beam is strongly absorbed and scattered by the plasma.

In our measurements, we recorded the incident pump beam, the transmitted pump beam, the broadband light emitted by the plasma, and the transmitted HeNe probe beam displayed on a screen.

IV. Experimental set-up

We used a single longitudinal mode, injection seeded, Q-switched YAG laser operating at 1064 nm. It is important to use a single longitudinal mode laser because its pulses have a well defined temporal shape which is free of mode beating. A multi-longitudinal mode laser pulse consists of numerous temporal spikes which makes the measurement the laser power at the instant of breakdown nearly impossible. For most of our measurements, we used a single laser pulse to damage the sample. To extract a single pulse or a number of pulses while keeping the Q-switched oscillator locked to the seed laser, we used an external shutter which was synchronized to the laser's Q-switch. We varied the pulse energy using a half wave plate and a high energy cube polarizer. The spatial filter consists of a 200 ⌈m diameter wire die followed by a variable iris which is adjusted to clip the transmitted beam at the first Airy null. The resulting beam is nearly TEM_{00}. We used fast phototubes (Hamamatsu, R1193U-51) to record the incident and the transmitted pulses, and a photomultiplier (RCA, IP28) to record the broadband breakdown light emitted from the focus. A HeNe probe beam was aligned co-linearly with the pump and displayed on a screen after passing through the sample. The sample was mounted on a motorized 3-axis translation stage, and we focused the pump beam into the sample using a 1-inch focal length, best form lens manufactured by CVI. Fig. 1 shows the schematics of our experimental set-up.

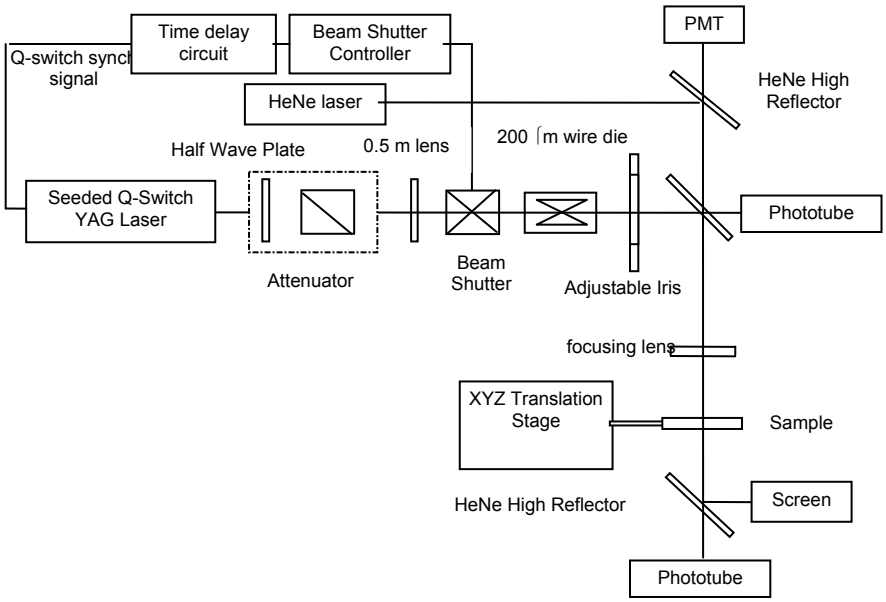


Fig. 1: Experimental set-up

V. Quality of laser pulses

Va. Temporal profile of laser pulses

The temporal profile of the 8 ns laser pulse is free of mode beating as may be seen by the profile displayed in Fig. 2. The laser pulses were highly reproducible with shot to shot pulse energy variation of approximately 1.5 %.

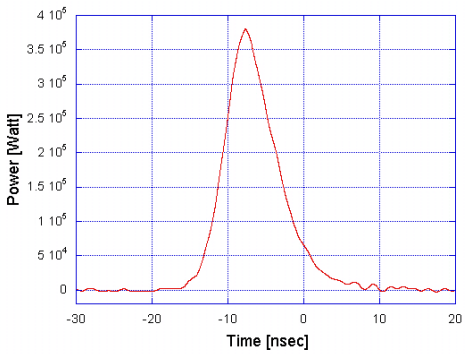


Fig. 2: The temporal profile of the Q-switched laser pulse; its FWHM is 8 ns

Vb. Spatial profile of laser pulses

The spatial profile of the nanosecond pulses before reaching the sample is shown in Fig. 3. Its profile is nearly Gaussian.

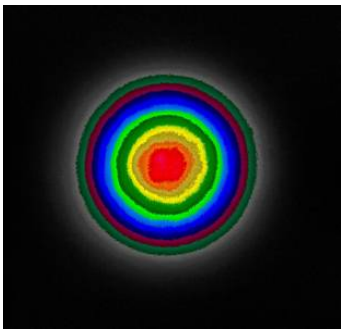


Figure 3: Spatial profile of the Q-switched laser beam before entering the sample

VI. The location and the size of the laser focus

We find the size and the location of the focus by measuring the surface third harmonic signal generated at the air-sample interface. This third harmonic signal is generated due to the broken symmetry at the air-solid interface [3]. This method is non-destructive, less time consuming, and more precise than the knife edge method; with the uncertainty of the focal position being less than 10 ⌈m (for 1" focal length lens). The third harmonic pulse energy for a focused lowest order Gaussian beam with a focal waist w_0 is proportional

$$U_{3\omega} = \frac{C_0}{w^4} = \frac{C_1}{\left(z_R^2 + \left[z - z_0\right]^2\right)^3}, \qquad (2)$$

where C_0 and C_1 are proportional constants, z_R is the Rayleigh range, and z_0 is the location of the window surface. The red dots in Fig. 4 are the measured third harmonic energy versus the *z* position of the sample and the blue solid line is the curve fitting of the experimental data using Eq. (2).

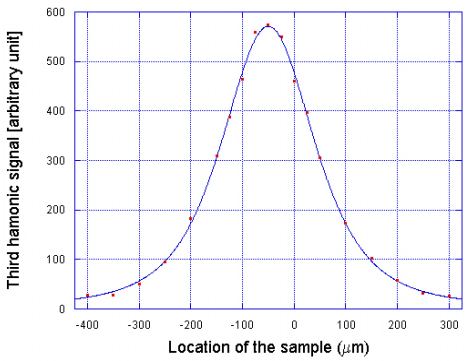


Fig. 4: Surface third harmonic signal as a function the location of the sample

The third harmonic is at its maximum when the beam waist lies on the sample surface. The measured third harmonic signal fits well with Eq. [2], and it shows excellent beam quality (M^2 = 1.0). The Rayleigh range, z_R is derived from the curve fitting and corresponds to a beam waist of 7.45 ⌈m. This agrees well with the value of the waist size measured by the knife edge method.

VII. Experimental results and discussion.

VIIa. Laser induced surface damage threshold of fused silica for nanosecond laser

We determined the surface damage threshold using the same method that we have successfully applied in measuring the bulk damage threshold [3]. We found the surface damage thresholds of Corning D1 grade fused silica windows prepared using the 3 different polishing methods described in part II. For single pulse threshold measurements we set the incident pulse energy approximately 25% above the bulk damage threshold. By matching the waveforms of the incident and transmitted laser pulses, the power at breakdown was determined from the point at which transmission through the fused silica window terminated, as illustrated in Fig. 5.

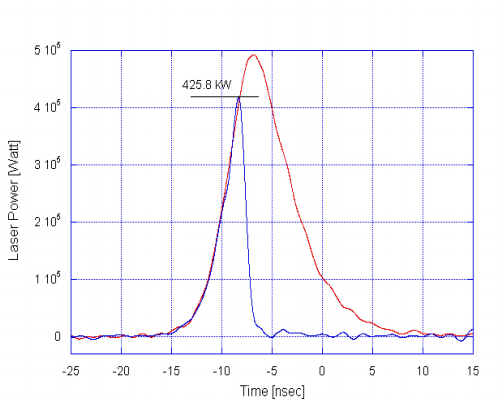
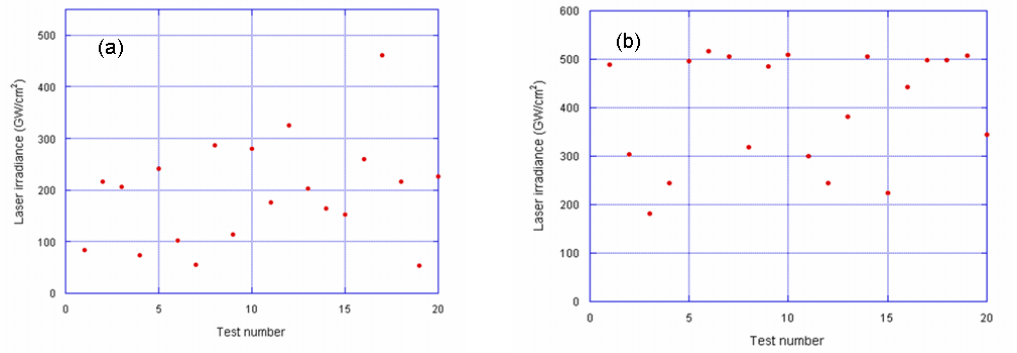


Fig. 5: Incident (red) and transmitted (blue) waveforms of surface damage, w_0 = 7.5 ⌈m, t_0 (FWHM) = 7.7 ns.

We found that the Cerium oxide polished sample showed damage at approximately 40% of the bulk threshold, with a large statistical spread (Fig. 6a), probably associated with the residual cerium oxide on the sample surfaces. The alumina polished sample showed damage between 50% and 100% of the bulk damage threshold (Fig. 6b). A substantial fraction of the measurements yielded the bulk value, and we believe it represents the intrinsic surface damage threshold. The large statistical spread might be due to contaminants or defects on the sample surface. This result suggested that a better alumina polish might achieve the intrinsic damage threshold at every test point. The alumina/silica polished sample achieves surface damage thresholds equal to the bulk damage value at nearly every point (Fig. 6c).



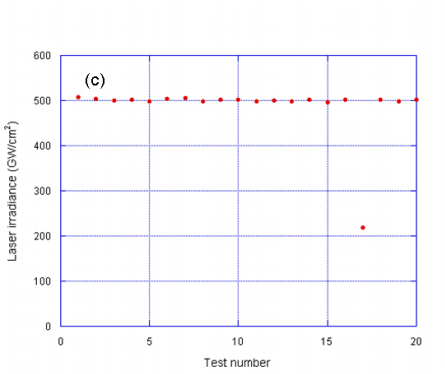


Figure 6: Single-shot surface damage threshold of (a) cerium oxide polished, (b) alumina polished, and (c) Alumina - Silica polished fused silica surfaces.

We concluded from these results that with the proper polishing process, the surface damage threshold of the input face of a fused silica window, can equal the bulk damage threshold, with little variation from point to point on the surface.

VIIb. Annealing effect on damage threshold of uncoated front surfaces polished with ceria and alumina

We also studied the effect of annealing on uncoated surfaces polished by ceria and by alumina, attempting to find a multiple-shot damage threshold. The laser pulse energy was raised from 0.36 to 4 mJ over three minutes (3.25 mJ is the bulk damage threshold). The laser beam was blocked after the first flash of white light in order to prevent extensive damage to the test window, and the pulse energy was measured. The multiple-shot damage threshold of the ceria polished surface was approximately 30% lower than its single-shot damage threshold, showing cumulative damage (Fig. 7a). The alumina polished surface showed the opposite effect, with most of the spots showing damage at the bulk damage threshold. The alumina surface showed a conditioning effect or a cleaning effect (Fig. 7b).

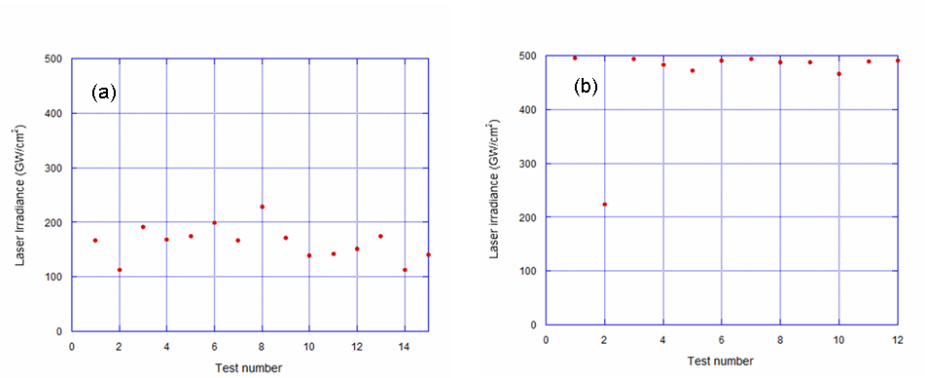


Figure 7: Multiple-shot surface damage threshold of (a) cerium oxide polished, (b) alumina polished fused silica surfaces.

VIIc. Focal size effect.

In the optical damage literature, it is sometimes claimed that the damage threshold irradiance (or fluence) depends on the size of the focal waist, with larger waists requiring lower irradiance. This is attributed to the increased number of defects covered by larger focal areas. The alumina/silica polished surface gives reliable surface damage thresholds, which are equal to the bulk damage thresholds, so we used it to test for a size effect. We used a 1" focal length lens, first positioning the focal waist on the front surface using the surface third harmonic signal, and then increasing the spot size by moving the window downstream. Fig. 8 shows the laser power at breakdown as a function of the distance from the front surface of the window to the focus spot. The equation of the fitting curve is

$$P(z=0)\left(1+\frac{z^2}{z_R^0}\right) \tag{8}$$

This shows there is no size effect for surface damage thresholds.

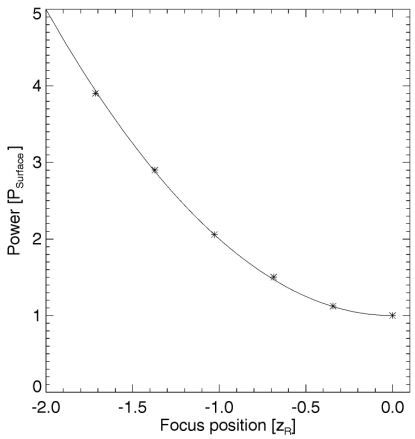


Fig. 8: Surface damage threshold laser power at different locations of the front surface.

VIId. Experimental study of self focusing effect in fused silica

Any bulk damage measurement must take into account, self focusing. The refractive index of fused silica may be approximated by

$$n = n_0 + n_2 I, \tag{3}$$

where n_0 is the low intensity refractive index of fused silica and n_2 is the nonlinear refractive index. n_2 has two contributions: an electronic Kerr effect and electrostriction. The Kerr contribution to n_2 for linearly polarized light is 2.2 x 10^{-20} m^2/W, and for circularly polarized light the Kerr contribution is 2/3 of that for linearly polarized light. The electrostriction contributes to n_2 in nanosecond regime is 0.5 x 10^{-20} m^2/W.

The self focusing power is given by

$$P_{sf} = \frac{0.149\lambda^2}{n\, n_2}. \tag{4}$$

The corresponding values of P_{sf} are 4.3 MW for linearly polarized light and 5.9 MW for circularly polarized light. The influence of self focusing on a Gaussian beam, when the power is below the self focusing power, is to move the focus down stream and decrease its focal waist. The correction factor for the maximum irradiance in the presence of self focusing depends on the instantaneous power and the depth of the focus inside the sample. The correction factor is plotted in Fig. 9 for different focal depths, which is measured in Rayleigh ranges, and for different powers measured in the unit of the self focusing power.

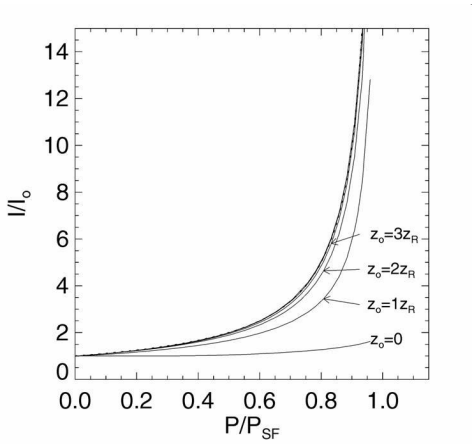


Figure 9: Irradiance correction factor for different depths of focus.

For focal depths of four or more Rayleigh ranges, the irradiance correction factor is

$$\frac{I_{corrected}}{I} = \frac{1}{1 - P/P_{sf}} \tag{5}$$

The self focusing correction is expected to be approximately 10% for the 8-⌈m focus when the focusing depth is more than 4 Rayleigh ranges. We can test our calculation of self focusing by varying the distance of the focus behind the input face of the sample. By using the alumina – silica polished fused silica window, we can find the surface damage threshold and bulk damage threshold at the focusing depth as small as 90 ⌈m reliably. The calculation of self focusing showed that the size of the focal waist decreased as the laser beam was focused deeper into the fused silica window. We expect to see the laser power at breakdown decreases as we moved the laser focus from the front surface to spots gradually deeper in the fused silica window. The results are shown in Fig. 10.

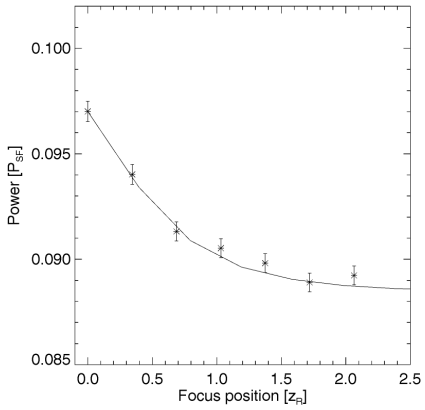


Fig. 10: Predicted (solid curve) and measured (symbols) damage threshold powers for different focusing depths behind the front surface, using a 1-inch focal length lens which gives an 8-⌈m focal waist.

Figure 10 shows the laser power at breakdown decreased as the laser focus spot was moved from the surface of the fused silica window to about 1.5 Rayleigh ranges behind the surface. The self focusing correction for the deepest focus is about 10%.

The correction factor for the location of the focus spot is a function of the laser power and its location under low field condition z_0 was illustrated in Fig. 11.

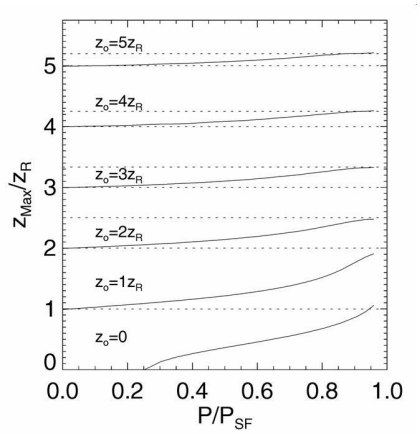


Fig. 11: The focal shift for different depths of focus at low field

Figure 11 shows that when the location of the focus spot is on the surface of the fused silica window under the low field condition. The focus spot remains on the surface of the silica window only when the laser power is smaller than 0.25 P_{sf}, when the laser power is greater than 0.25 P_{sf}, the laser focus spot moves into the bulk behind the window surface. This is the reason for keeping the laser power at breakdown well below 0.25 P_{sf} by keeping the laser focus spot small.

VIII. Damage morphology

With the pulse energy set 25 % above the bulk damage threshold we studied the single-shot damage morphology and found a physical difference in morphologies between spots on the alumina-silica polished surface that damaged at the bulk damage threshold and those on the Cerium oxide polished surface that damaged well below the bulk damage threshold. In the former, the damage consisted of multiple radial fractures similar to the end view of the bulk damage (Fig. 12a); in the latter, there was no

fracturing (Fig. 11b). For example, Fig. 12b shows damage generated at 40% of the bulk damage threshold. This picture was taken by a phase contrast microscope and it shows a change in the refractive index at the damaged spot. Optical breakdown generated at low powers shields the fused silica surface from the rest of the laser pulse and so keeps the damage small. Figure 12c shows a phase contrast image of damage generated at 65% of the bulk damage threshold. Damage appears as a blue dimple with surrounding rings. The blue dimple can easily be observed using a normal microscope, but the rings cannot.

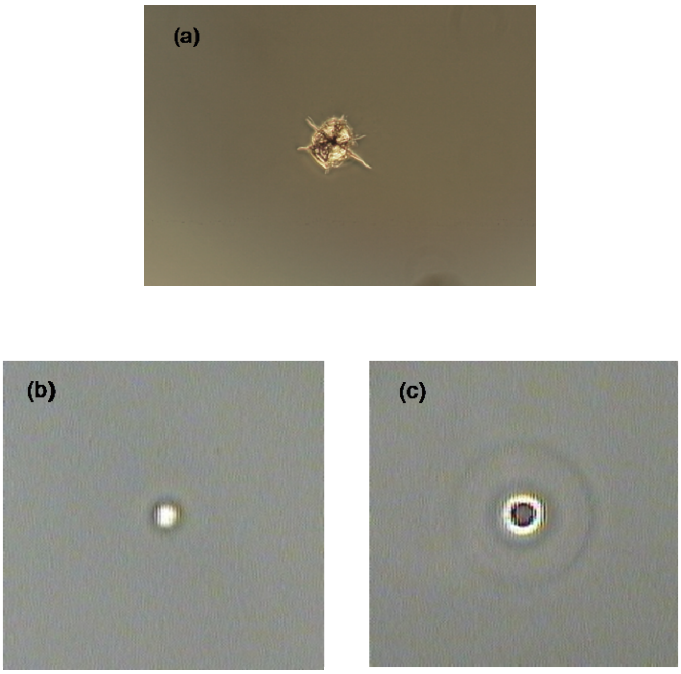


Figure 12: Surface optical damages generated at (a) 10% above the bulk damage threshold power, (b) ~ 40% of the bulk damage threshold power, (c) ~ 65% of the bulk damage threshold power.

IX. Conclusions

We have shown that the single-shot surface damage threshold of ceria polished silica is statistical and well below the bulk damage threshold. Its multiple-shot damage threshold is even lower than the single-shot threshold, which indicates cumulative damage. The single-shot surface damage threshold of alumina polished silica is also statistical and lower than its bulk damage threshold. However, its multiple-shot damage threshold is deterministic and equal to the intrinsic bulk damage threshold, indicating a

conditioning effect. The single-shot surface damage threshold of the alumina/silica polished surface is deterministic and equal to the bulk silica damage threshold.

By using a fused silica window polished by alumina/silica, we were able to show that the laser irradiance at breakdown is independent of the focus size. By measuring laser power at breakdown at different focusing depths, we showed experimentally, the self focusing effect in fused silica. In order to obtain the correct optical damage threshold irradiance or fluence in the bulk, the self focusing correction must be taken into account. Because of this self focusing effect, the size of the focus spot in the damaging process is smaller than the focus spot under a low field condition.

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How to polish fused silica equal to obtain surface damage threshold equal to the bulk damage [7842-85]

Questions and Answers

Q. So, at the beginning of your talk, you said there is damage when you have a contaminant or when you have a crack. You referenced Bloembergen and said that damage will initiate when you have contamination present or a crack that initiates intensification.

A. No, I didn't say that. What I said was, I quoted from him that if you have a defect, right, then the actual defect that you have will affect the field enhancement and it will go by the square of the refractive index. The damage threshold can go down by a factor of the fourth power of the refractive index. So, we can reduce the damage threshold, or optical breakdown threshold. This doesn't mean that you have to have a defect in order to generate breakdown damage. If you reach a critical intensity, then you have it.

Q. It's a question of definition. All right, let me move on. So then you do it on twenty sites using your beam size which you haven't defined, which I assume is on the order of one micron.

A. The radius of the beam is 7.45 micron at $1/e^2$.

Q. So, basically what you're testing is to see whether you have any of those causes of damage within the small volume that you tested. So, basically you do 20 shots over 20 micron areas which is a very small amount of surface to test. And, if you get damage, it means, even in the best case, you have one of those things in that volume.

A. Yes. I do many sets of measurements. I don't just do one and go home and sleep. No. I do many sets, and that one set may show damage. Sometimes I do 18 out of 20.

Q. So now, most of the optical elements are of much larger size, and therefore, with those kinds of statistics you're going to have a very large number of those initiation sites. Therefore, even if you use the best data you have here, you still don't have a good optic.

A. It's better than the conventional one. Is that right?

Q. If you can explain this, I will appreciate it.

A. Let's say I look at this one, the one for the alumina polish, then this one is the conventional one, right. This one is polished by alumina. There already is an improvement. Certainly, I agree with you because, we have, let's say 5% defect from this type of polishing. I agree.

Q. I just wanted to expand on Stavros' comment. For example, the process described in a previous talk, the projected number of damage sites on a NIF-size optic is two. If I understand your test correctly, it seemed like you measured a square mm of area. Is that correct?

A. More than that. If you take the area and multiply, then you have a small area. I shoot a shot then move a couple of millimeters away and shoot another shot and so on. So, I can sample a bigger area.

Q. The crux of my point is that it has been shown as recently as yesterday, that spot size has a dramatic effect on damage threshold. So, I just caution coming to conclusions about surface quality by sampling very small areas of the surface.

A. Yes. But the reason is I have to use a small laser beam because I want to keep the laser power at breakdown to be lower than 20% of the critical power for self-focusing. If I increase the area by a factor of two, then practically, the area goes by the square of the radius, right? Then, the power at breakdown is bigger than 25% of the critical power. Then, the laser focus spot is not on the surface, it moves in to the part. That's why the spot size needs to be small. That's the reason for that. I

understand, but because the power at breakdown in my case is already 10% of the critical power, I cannot increase the spot size because, I will go past the surface. If I increase the spot size by a factor of two, I will get more. I don't get just the surface.