

Rate equation model of bulk optical damage of silica, and the influence of polishing on surface optical damage of silica

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Abstract

Our objective is to understand the mechanism that generates catastrophic optical damage in pulsed fiber amplifiers. We measured optical damage thresholds of bulk fused silica at 1064 nm for 8 ns and 14 ps pulses. The 8 ns pulse is single longitudinal mode from a Q-switched laser, and the 14 ps pulse is from a Q-switched mode-lock laser. The beams in both cases are TEM₀₀ mode, and they are focused to a 7.5 μm spot inside a fused silica window. The pulse-to-pulse energy variations are 1% for 8 ns pulses and 5% for 14 ps pulses. Under these conditions optical damage is always accompanied by plasma formation at the focal spot; we found the damage threshold fluences are $3854 \pm 85 \text{ J/cm}^2$ for the 8 ns pulses and $25.4 \pm 1.0 \text{ J/cm}^2$ for the 14 ps pulses. These fluences are corrected for self focusing. Both damage thresholds are deterministic, in contrast to the claim often made in the literature that optical damage is statistical in the nanosecond range.

The measured damage threshold fluences for 8 ns and 14 ps pulses do not fit a square root of pulse duration scaling rule. We interpret the damage in terms of plasma formation initiated by multiphoton ionization and amplified by an electron avalanche. The damage threshold irradiance can be matched with a simple rate equation model that includes multiphoton ionization, electron avalanche, and electron-hole recombination.

The damage morphologies are dramatically different in the nanosecond and picosecond cases because of the large difference in deposited energy. However, both morphologies are reproducible from pulse to pulse.

We also measured surface damage thresholds for silica windows polished by different methods. We find that cerium oxide polished surfaces damage at approximately 40% of the bulk threshold, with a large statistical spread. Surfaces prepared using an Al₂O₃ polish damaged between 50% and 100% of the bulk damage limit, with a substantial fraction at 100%. Surfaces polished using first the Al₂O₃ polish and then an SiO₂ polish exhibit surface damage values equal to the bulk damage value at nearly every point.

We also measured damage thresholds for different sized focal spots. Some earlier reports have claimed that damage thresholds depend strongly on the size of the focal spot, but we find the surface threshold is independent of the spot size.

I. Introduction

Laser induced breakdown leading to optical damage in optically transparent material such as fused silica is important in the development and application of high power lasers because it imposes a limit on the system performance. However, the reported values of

damage threshold irradiance/fluence cannot be used as a basis for fiber amplifier design because they vary over orders of magnitude. For example, in 1980, M. Soileau and M. Bass reported the damage threshold irradiance of fused silica was 605 GW/cm^2 for a pulse duration of 31 ns and a beam radius of $6.15 \text{ }\mu\text{m}$ [1]. More recently Natoli *et. al.* reported the damage threshold irradiance of fused silica was $44 \pm 12 \text{ GW/cm}^2$ for a pulse duration of 7 ns, with a beam radius that varied from 10 to $100 \text{ }\mu\text{m}$ [2]. We believe better measurements of the optical breakdown threshold and a better understanding of the optical breakdown process are called for.

In our studies of optical breakdown, we address a number of questions and measurement issues:

1. Is the optical breakdown threshold 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?
4. Is the breakdown threshold irradiance independent on the size of the focus spot?
5. Does the breakdown threshold scale as the square root of the pulse duration?
6. What is the role of Stimulated Brillouin Scattering (SBS)?
7. What is the role of self focusing?
8. What is the effect of mechanical stress on the bulk breakdown threshold?

II. Experimental set-up and technique

1. Detection of optical breakdown

Intense laser light excites electrons from the valence band to the conduction band by three processes: tunneling ionization, multiphoton ionization, 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} ,

$$\omega_p^2 = \frac{e^2 n_{crit}}{m^* \epsilon \epsilon_0} = \omega^2, \quad (1)$$

where e is the electron charge, m^* is the effective electron mass, ϵ is the relative permittivity of the medium, and ϵ_0 is the vacuum permittivity. For 1064 nm light, the critical density of fused silica is $2.08 \times 10^{21}/\text{cm}^3$. When the free electron density reaches this level, incoming laser light is strongly reflected. However, the energy deposited in the free electrons at this density is sufficient to cause permanent damage through melting and fracturing. At the onset of optical breakdown the following processes occur [3]:

- 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 that causes 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.

2. Experimental set-up

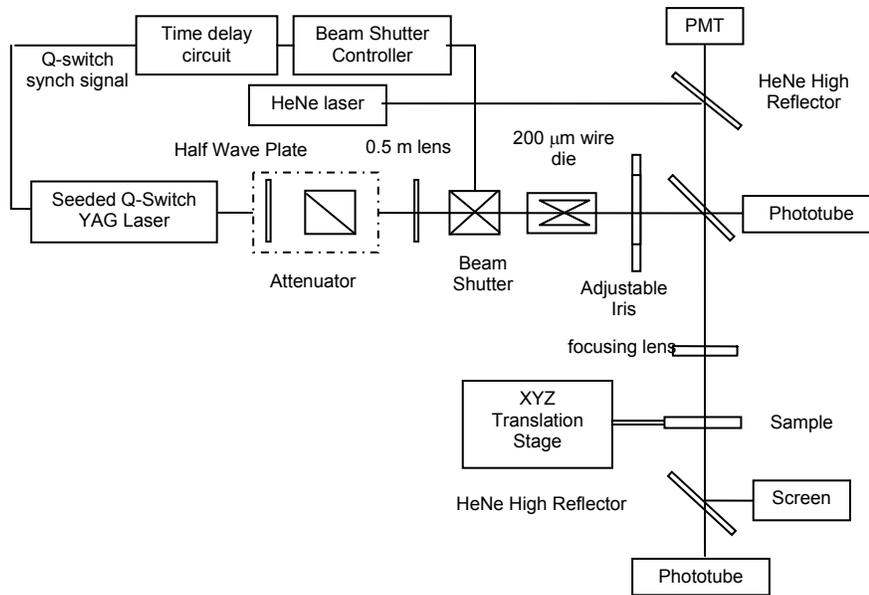


Figure 1: Experimental set-up

Figure 1 shows our measurement apparatus. Damage is created by laser pulses from a single longitudinal mode, injection seeded, Q-switched YAG laser. It is important to use a single longitudinal mode laser because its pulses have a well defined temporal shape free of mode beating. A multi longitudinal mode laser pulse consists of numerous temporal spikes, making a measurement of the power at the instant of breakdown impossible. For most of our measurements, we used a single laser pulse to damage the sample. To extract a single pulse or a limited number of pulses and still keep the Q-switched oscillator locked to the seed laser, we used an external shutter synchronized to the 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 μm diameter wire die followed by a variable iris which is adjusted to clip the beam at the first Airy null that filters the spatial mode, making it nearly TEM_{00} . We used fast phototubes (Hamamatsu, R1193U-51) to record the incident and the transmitted pump beams, and a photomultiplier (Hamamatsu, R406) to record the broadband breakdown light emitted from the focus. A HeNe probe beam was aligned coaxially 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 1" focal length best form lenses manufactured by CVI.

3. Time and spatial profiles 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 figure 2. Pulses were highly reproducible with shot to pulse energy variations of approximately 1%.

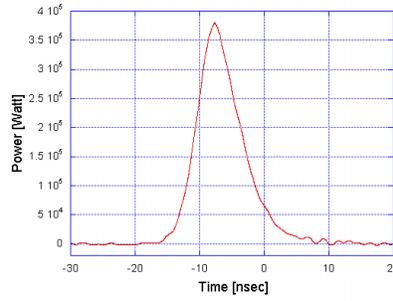


Figure 2: The temporal profile of the Q-switched laser pulse; its FWHM is 8 ns

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

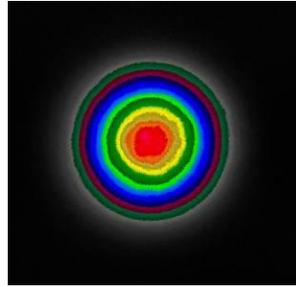


Figure 3: Spatial profile of the Q-switched laser beam before entering the sample

The spatial profile of the 14 picosecond pulses is also TEM₀₀; the pulse energy varies by approximately 5% from pulse to pulse.

4. Location and the size of the laser focus

We can 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 was generated due to the broken symmetry at the air-solid interface [4]. This method is non-destructive, less time consuming, and more precise than the knife edge method; the uncertainty of the focal position is less than 10 μm (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 to

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

where C is a proportional constant, z_R is the Rayleigh range, and z_0 is the location of the window surface. Figure 4 shows the measured third harmonic energy versus the z position of the sample is fitted to a line of Eq. (2).

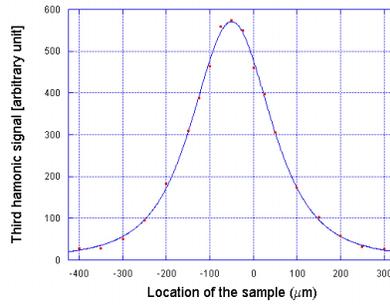


Figure 4: Surface third harmonic signal as a function the z position of the sample

The third harmonic is maximized when the beam waist lies on the sample surface. The measured signal fits well with equation (2) which verifies excellent beam quality ($M^2=1.0$). The Rayleigh range, z_R is derived from the curve fit corresponds to a beam waist of $7.45 \mu\text{m}$ for the $1''$ focal length lens. This agrees well with the value of the waist size measured by the knife edge method.

5. Theoretical calculation of self focusing

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

$$n = n_0 + n_2 I, \quad (3)$$

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 $2.2 \times 10^{-20} \text{ m}^2/\text{W}$, 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 $0.5 \times 10^{-20} \text{ m}^2/\text{W}$, and it does not contribute to n_2 in the picosecond regime because the time is too short for any material to move $8 \mu\text{m}$ (laser focus spot size) in fused silica, since the longitudinal sound velocity in silica is 6000 m/sec and the time for the sound to travel $8 \mu\text{m}$ is 1.3 ns .

The self focusing power is given by

$$P_{sf} = \frac{0.149 \lambda^2}{n n_2}. \quad (4)$$

The corresponding values of P_{sf} are 4.3 MW (linear polarization, 8 ns pulses), 5.9 MW (circular polarization, 8 ns pulses), 5.3 MW (linear polarization, 14 ps pulses), 7.9 MW (circular polarization, 14 ps pulses). 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 Figure 5 for different focal depths, which is measured in Rayleigh ranges, and for different powers measured in the unit of the self focusing power.

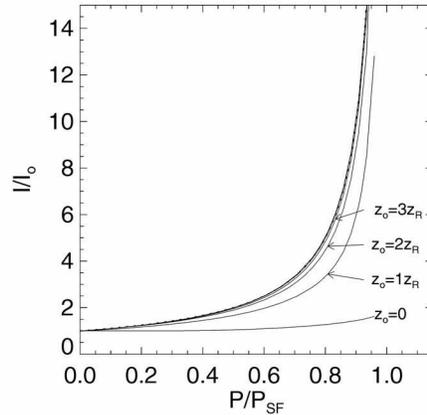


Figure 5: Irradiance correction factor for different depths of focus.

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

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

6. Stimulated Brillouin Scattering (SBS).

SBS is another effect that can influence damage measurements. It can reflect a phase conjugated beam that interferes with the incoming beam to create antinodes of high irradiance, thus lowering the apparent damage threshold. Alternatively, SBS can reflect the incoming beam before it reaches the focus, raising the apparent threshold. In either situation, it should be avoided if possible. Experimentally, we found the SBS threshold in D1 fused silica for linearly or circularly polarized light to be 0.85 MW. Figure 6 shows incident, transmitted, and amplified SBS beams. The pulse reflected by SBS propagated back into the laser and was amplified before returning to the sample; therefore it reappeared about 50 ns after the main pulse and emerged as the delayed weak pulse in the oscilloscope trace.

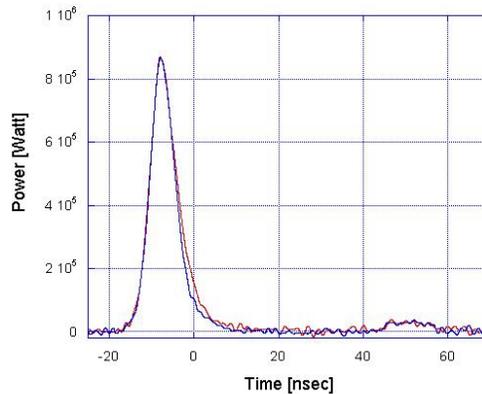


Figure 6: Incident (red) and transmitted (blue) waveforms of a pulse slightly above the SBS threshold. $w_0 = 17\mu\text{m}$, $t_0(\text{FWHM}) = 7.5\text{ns}$.

The SBS threshold power is nearly independent of the focal waist size. For reliable damage measurements we must use conditions that avoid SBS. This requires focal sizes of 15 μm or less.

III. Experimental results and discussion.

1. Laser induced damage threshold in bulk fused silica for seeded nanosecond laser

We used 1" focal length best form lens and by measuring the surface third harmonic signal we set the focus spot 2.9 mm behind the sample surface. This is more than 10 times the Rayleigh range of 238 μm , ensuring that we did not have surface damage. We used a single laser pulse at different locations in the sample, gradually increasing the pulse energy at each location until optical breakdown occurred. The pulse energy difference between 100% and 0% damage probability was less than 2.0%. This sharp damage threshold was independent of spot location across the samples, and different fused silica types (Corning A0, B0, B1, D0, D1, D3, D5) gave identical results.

Figure 7 shows the transmitted pulse profiles for different pulse energies; below and above the damage threshold. The power at breakdown increased about 5%, as the laser pulse energy increased from the damage threshold to more than 1.5 times of that value. This shows a nonzero induction time of less than one nanosecond. Breakdown occurs at a nearly fixed irradiance rather than a fixed fluence. The threshold power is 374 kW for linearly polarized light and 387 kW for circularly polarized light, corresponding to irradiances of $475 \pm 7\% \text{ GW/cm}^2$ for both polarizations after correcting by approximately 10% for the effect of self focusing.

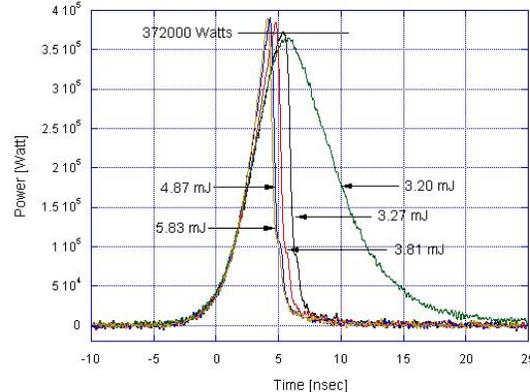


Figure 7: Transmitted waveforms for pulse energies below and above the optical breakdown threshold, $w_0 = 7.45 \mu\text{m}$, t_0 (FWHM) = 7.5 ns.

2. Laser induced damage threshold in stressed fused silica

Polarization maintaining fibers use built in strain to maintain polarization. The strain induced birefringence typically gives Δn of approximately 10^{-4} . Therefore, we measured the damage threshold at this strain level to see whether it is altered by strain. We simulated this strain level by compressing a silica brick in a press. Our brick was a 1.27 cm x 1.27 cm x 1.27 cm cube of fused silica. It was compressed to produce a half wave of retardation difference between the two eigen polarizations at 633 nm at the center of the

brick. This corresponds to a birefringence of Δn of 2.5×10^{-5} , comparable to that in a polarization maintaining fiber. Damage thresholds were measured for both polarizations. The two damage thresholds were equal, and they were identical to the damage threshold of the unstressed fused silica within our experimental uncertainty of 2%.

3. Laser induced damage threshold in bulk fused silica for picosecond laser

We also measured optical damage thresholds of bulk fused silica at $1.064 \mu\text{m}$ for 14 ps pulses from a Q-switched mode-lock laser. The output of this laser was spatially filtered to produce a nearly TEM_{00} beam that was focused to a $7.5 \mu\text{m}$ waist located 2.9 mm behind the window surface. The pulse to pulse energy variation is 5%. We found the damage threshold fluence was $25.4 \pm 1.3 \text{ J/cm}^2$. This is corrected for self focusing which reduces the focal waist area by 27%.

4. Pulse duration scaling of damage threshold in bulk fused silica

The literature on optical damage in transparent material often claims that the damage threshold fluence scales as the square root of the pulse durations ($t^{1/2}$) in the ps-ns range. However, the ratio of the square root of the pulse durations for our 8 ns and 14 ps pulses is 24, but the ratio of our two measured damage threshold fluences at these two pulse durations is 152. Under the conditions of our measurements the square root of the pulse duration scaling is clearly inaccurate. The $t^{1/2}$ behavior is sometimes claimed to support a thermal initiation of damage. However, we interpret the optical damage in our measurements as resulting from plasma formation generated by multiphoton ionization and electron avalanche. The laser energy deposited in the free electron gas at critical density is sufficient to generate optical damage. The damage threshold irradiance is weakly dependent on the pulse duration as we showed in sections III.1 and III.3. The damage threshold fluence in fused silica in the ps-ns range can be predicted by a rate equation model; which will be explained in section III.9.

4. Damage morphology

The bulk damage morphology in silica is quite reproducible from pulse to pulse. Figure 8 shows a side view of several damage spots created by single longitudinal mode nanosecond pulses whose energy just slightly above the damage threshold.

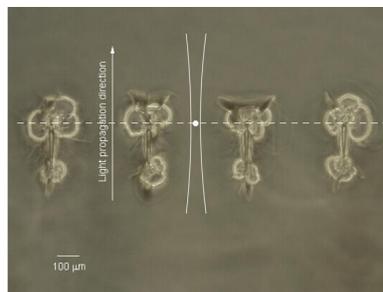


Figure 8: Side view of optical damage spots in fused silica. The pulse energy was slightly above the damage threshold.

The dashed line shows the location of the focal waist, and the size of the dot indicates the uncertainty ($\pm 10 \mu\text{m}$) in waist depth. The two curved lines indicate the beam size one Rayleigh range either side of the focus. Our interpretation of the damage is that optical breakdown starts at the exact waist where the laser intensity is highest (the self focus shift in the waist location is negligible here). After damage is initiated, the damage grows upstream as energy is absorbed from the incoming pulse. After propagating upstream approximately one Rayleigh range, the irradiance is insufficient to sustain the plasma and the damage growth ceases.

The optical damage generated by picosecond pulses is much smaller than that generated by nanosecond pulses because of the difference in pulse energies. Optical damage in the picosecond regime is also reproducible.

5. Nanosecond laser induced surface damage threshold of fused silica

By measuring the surface third harmonic signal, we can place the focal waist exactly on the front surface of the sample so the irradiance is precisely known. We measured the surface damage thresholds of D1 silica windows prepared using three different polishing methods. The first was a cerium oxide polish. This type of polish is common in commercially available windows. The second was an alumina polish performed at Alpine Research Optics (ARO). All polishing steps used alumina polishing compound with the final polish being 100 nm alumina. The third was the same as the second window except it was further polished using 40 nm silica. We set the incident pulse energy about 30% above the bulk damage threshold and determined the damage threshold from the point of transmission cut off.

We find that the ceria polished window damaged at approximately 40% of the bulk threshold, and has a large statistical spread, probably associated with the residual cerium oxide incorporated into the polished surface. The alumina polished window damaged between 100% and 50% of the bulk damage limit, but a substantial fraction of the measurements yield the bulk value, and we believe it represents the intrinsic surface damage threshold. The large statistical spread might be due to contaminants and defects on the sample's surface. The silica polished window exhibited surface damage values equal to the bulk damage value at nearly every point. These results are shown in figures 9, 10 and 11.

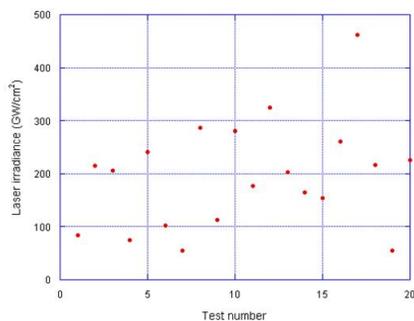


Figure 9: Surface damage threshold of Cerium Oxide polished surface.

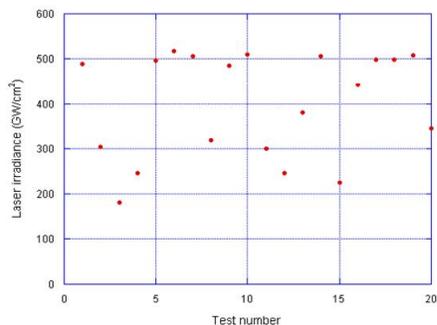


Figure 10: Surface damage threshold of Alumina polished surface.

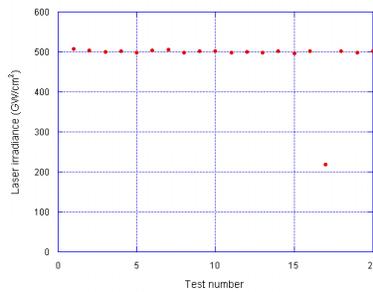


Figure 11: Surface damage threshold of Alumina - Silica polished surface.

We surmise that with a proper polishing process, the input surface damage threshold for fused silica is equal to the bulk damage threshold with little variation from point to point on the surface.

7. 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, which was attributed to a larger number of defects included in a larger focal volume. The silica polished surface gives a reliable surface damage threshold, that is equal to the bulk damage threshold, so we used it to test for a size effect. We used the 1" focal length lens and set the focus spot on the front surface of the silica window by maximizing the surface third harmonic. To increase the spot size on the surface we move the window away from the focus spot by a known distance. We found the laser power at breakdown by noting the point of transmission cut off. Figure 12 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

$$P = P(z = 0) \left(1 + \frac{z^2}{z_R^2} \right) \quad (6)$$

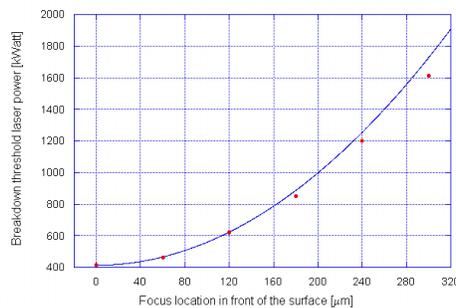


Figure 12: Surface damage threshold laser power at different locations of the front surface. Dots are measured and the curve has the form of Eq. (8).

Within the uncertainty of our measurement the damage threshold irradiance is independent of size for focal spots from 7 to 15 μm .

7. Electron avalanche model for nanosecond and picosecond pulses

An important question is whether an electron avalanche model can account for the damage in silica. The simplest electron avalanche rate model is described by the electron density rate equation [5]

$$\frac{dn}{dt} = \beta I^8(t) + \alpha n I(t) - n/\tau \quad (7)$$

where n is the electron density, β is the rate for the 8 photon absorption necessary for 1064 nm ionization of silica, α is the electron avalanche coefficient, and τ is the free electron lifetime due to electron-hole recombination. The electron lifetime in fused silica is approximately 150 fs [6] according to numerous femtosecond studies. Optical breakdown requires that the avalanche term be large enough to overcome the large recombination term. Once this happens the avalanche to the damage point is nearly instantaneous on the time scale of our 7.5 ns pulses. The requirement that the avalanche term $\alpha n I(t)$ exceed the recombination term n/τ , implies that $I(t)$ must exceed $1/\alpha\tau$ for optical breakdown. The bulk damage threshold irradiance (nanosecond) for fused silica was found to be 475 GW/cm². The electron avalanche model matches our optical breakdown thresholds reasonably well, provided $\alpha\tau = 2 \times 10^{-4} \mu\text{m}^2/\text{W}$.

The values of $\tau = 250$ fs and $\alpha = 6.1 \times 10^8 \mu\text{m}^2/\text{J}$ gave a good fit to the measured thresholds of the 8 ns and 14 ps pulses as shown in figure 13.

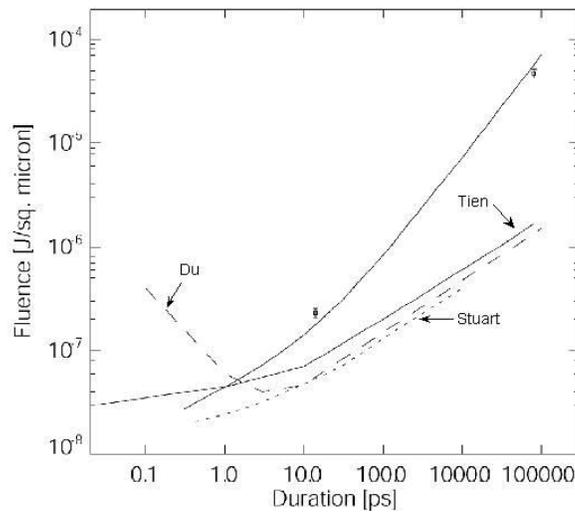


Figure 13: Damage threshold fluence of fused silica in the picosecond to nanosecond regime as predicted by the rate equation (solid line) and our two data points for 8 ns and 14 ps pulses. The labeled curves show measurements of surface damage made by other researchers using larger diameter beams.

The value of β we used in our simulation is $1 \times 10^{-21} \mu\text{m}^{13}/\text{sW}^8$. It is in reasonable agreement with rates measured using femtosecond pulses [7] for 8 photon absorption of 1.064 μm light.

IV. Conclusions

We report that the bulk damage threshold irradiance in fused silica is $475 \times 10^9 \pm 7\%$ Watts/cm² for linearly or circularly polarized light, a pulse duration of 7.5 ns, and a focal waist of 7.45 μm . The damage threshold irradiance of stressed fused silica is the same as the unstressed one. The surface damage threshold is nearly the same as the bulk damage threshold if the sample's surface is properly prepared.

The stimulated Brillouin threshold power in fused silica is 851.0 ± 2.9 kW for linearly or circularly polarized tightly focused beams at our pulse duration of 7.5 ns. Below the SBS threshold, we showed that the damage threshold irradiance is independent of the laser beam spot size. Above the SBS threshold the damage threshold irradiance is perhaps reduced due to the inference between the incident pulse and the SBS pulse. We showed that an electron avalanche described by a simple rate equation can successfully fit our measured breakdown thresholds in fused silica over the nanosecond and picosecond range.

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