# Deterministic Nanosecond Laser-Induced Breakdown Thresholds In Pure and Yb<sup>3+</sup> Doped Fused Silica

#### Arlee Smith, Binh Do

Laser, Remote Sensing, Plasma and Complex System Science Department Sandia National Laboratories, Albuquerque, New Mexico

## Mikko Soderlund

Liekki Corporation, Lohja,Finland

## Abstract:

The objective of this work is to understand catastrophic optical damage in nanosecond pulsed fiber amplifiers. We used a pulsed, single longitudinal mode, TEM<sub>00</sub> laser at 1.064  $\mu$ m, with 7.5-nsec pulse duration, and focused it to a 7.45- $\mu$ m-radius spot in bulk fused silica. The bulk damage threshold irradiance was corrected by approximately 10% to account for self focusing. The pulse to pulse variation in the damage irradiance in pure silica was less than 1%. The damage was nearly instantaneous with an induction time much less than 1 ns. These observations are consistent with an electron avalanche rate equation model with reasonable rate coefficients. The bulk optical breakdown threshold irradiance of pure fused silica is  $4.7 \times 10^{11} \pm 3\%$  Watts/cm<sup>2</sup>. We also measured the surface damage threshold irradiance of 1% Yb<sup>3+</sup> doped fused silica preform of Liekki Yb1200 fiber, and found it is equal to that of pure silica within 2%.

The optical damage morphology is reproducible from pulse to pulse. To facilitate the morphology study we developed a technique for locating the laser focus based on the third harmonic signal generated at the air-fused silica interface. This gives a very small uncertainty in focal position (~ 10  $\mu$ m) which is important in interpreting the damage structure. The surface third harmonic method was also used to determine the laser focus spot size.

Earlier reports have claimed that the damage irradiance depended strongly on the size of the focal spot. We varied the focal volume to look for evidence of this effect, but found no evidence for it.

#### I. Introduction:

Laser induced breakdown leading to optical damage in optically transparent material such as fused silica has been studied by many researchers since the invention of laser more than four decades ago. It has been important in the areas of development and applications of high power lasers because laser induced damage threshold was an ultimate limit to system performance. However, the literature on nanosecond damage of fused silica was too confusing to use as basis for fiber amplifier design, the reported values of damage threshold irradiance/fluence vary over orders of magnitude. For example, in 1980, M. Soileau and M. Bass reported that the damage threshold irradiance of fused silica is 605 GW/cm<sup>2</sup> for the pulse duration of 31 ns, and the beam radius at  $1/e^2$  was 6.15  $\mu$ m [1], more recently L. Gallais et al reported that the damage threshold irradiance of fused silica was  $22 \pm 5$  GW/cm<sup>2</sup> for the pulse duration of 7 ns, and the beam

radius at  $1/e^2$  was 6 µm [2]. We need better measurements of the optical breakdown threshold and understanding of the optical breakdown process. We also need to measure the damage threshold of Yb<sup>3+</sup>-doped fused silica. In addition, we wanted to find out whether the optical breakdown threshold is determined by irradiance or fluence.

When we were looking for optical breakdown threshold, we addressed a number of issues:

- 1. Detection of optical breakdown.
- 2. Time and spatial profiles of laser pulses.
- 3. Location and the size of the laser focus.
- 4. Self focusing.
- 5. Stimulated Brillouin Scattering (SBS).

After that we were ready to find the bulk optical breakdown damage threshold, to study the damage mechanism and morphology, the focal size effect, and the influence of  $Yb^{3+}$  doping in fused silica.

## **II. Experimental set-up and technique.**

## 1. Detection of optical breakdown.

Laser excites electrons into the conduction band by three processes: Tunneling ionization, multiphoton ionization, impact ionization. When the free electron's density reaches the critical density, then the plasma frequency is equal to the laser frequency.

$$\omega_{\rm p}^2 = \frac{{\rm e}^2 n}{{\rm m}\varepsilon\varepsilon_0} = \omega_{laser}^2 \tag{1}$$

Where e is the electron charge, n is the free electron density, m is the electron mass,  $\varepsilon$  is the relative permittivity of the medium and  $\varepsilon_0$  is the free space permittivity. We used 1.064 µm laser, the corresponding critical densities of free electrons in fused silica is 2.08 x  $10^{21}$ /cm<sup>3</sup>.

When the optical breakdown begins, the following observations occur at the same time [3].

a. The generation of a high-density plasma at the focusing region of the laser pulses that appears as a bright white spot accompanied by the emission of a broadband light. We detect the white light emitted by this plasma as the primary indicator of optical breakdown.

b. The drastic decrease in the transmitted laser power as the incident laser pulse is absorbed, reflected and scattered by the dense plasma created by the optical breakdown at the focal region.

c. The decrease in the power of the transmitted HeNe probe beam. At the onset of the optical breakdown, the dense plasma starts to scatter the probe beam.

In our experimental set-up, we had detectors to record the incident pump beam, transmitted pump beam, the broadband light generated by the optical breakdown region and a screen to display the transmitted probe beam.

#### 2. Experimental set-up.



# Figure 1: Experimental set-up

In this experiment, we used a single longitudinal mode injection seeded Q-switch YAG laser. It was very important to use a single longitudinal mode laser in this experiment because its laser pulses have well defined temporal shape. In contrast, if we used a multiple longitudinal mode laser, the mode beating problem would made the laser pulse consisted of numerous temporal spikes, then the measurement of laser power at breakdown would become very unreliable or impossible. Most of the time, we used a single laser pulse from our laser and tried to damage our sample, the laser could run in the single shot mode, however, the host oscillator could not locked to the frequency of the seed laser in this running mode. In order to extract a singe laser pulse or a number of laser pulses and to keep the host oscillator locks to the frequency of the seed laser, we used a beam shutter which used the synch Q-switch signal, it opened at 40 ms before the first pulse and closed at 40 ms after the last pulse. We varied the pulse energy by using an attenuator which consists of a half wave plate and a high energy cube polarizer. We used a 200 µm wire die and adjustable iris to clean up the spatial profile of the laser beam and made it close to TEM00 mode. We also used two phototubes (Hamamatsu, R1193U-51) to record the incident and the transmitted pump beams, and a photomultiplier (Hamamatsu, R406) to record the broadband light emitted from the optical breakdown region. The transmitted HeNe probe beam which was sent collinearly with the pump and it was displayed on a screen after going through the sample. The sample was mounted on a motorized 3-dimensional translation stage and we focused the pump beam into the sample by using 1", 1.5", and 2" focal length best form lenses manufactured by CVI.

## 4. Time and spatial profiles of laser pulses.

It was very important to have a smooth temporal profile for our laser pulses, our injection seeded laser produced single longitudinal mode beam, its temporal profile was smooth and it was shown in figure 2. It was very reproducible, its energy varied about one percent from shot to shot.



Figure 2: The temporal profile of our laser beam, its FWHM is 7.5 ns.

In order for the laser beam focus properly, its spatial profile should be very close to TEM00 mode, figure 3 showed the spatial profile of our laser pulses before it went into the sample.



Figure 3: The spatial profile of our laser beam before going into the sample. **5. Location and the size of the laser focus.** 

In this experiment we wanted to measure the laser power at breakdown and we also wanted to study the damage morphology generated by the optical breakdown. This study was precise only if we knew exactly the location of the focusing spot and the dimension of the focusing spot size in our sample. We could perform these two tasks by measuring the surface third harmonic signal generated by the air-solid interface. This third harmonic signal was due to the broken symmetry of the air-solid interface. The bulk third harmonic signal did exist but it was very much weaker than the surface third harmonic signal [4]. This method was non-destructive and the uncertainty was less than 10  $\mu$ m (for 1" focal length lens). The surface third harmonic pulse energy was proportional to

Energy<sup>(3
$$\omega$$
)</sup> =  $\int_{0}^{\infty} \int_{0}^{2\pi} I_{\omega}^{3} \exp\left(-6\frac{r^{2}}{w^{2}}\right) r d\theta dr \approx \frac{1}{w^{4}}$  (2)

Where w was the beam radius on the surface of the sample. Figure 4 showed the surface third harmonic signal with respect to the nominal location of the sample.



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

Thus, when the surface third harmonic signal was maximum then the beam waist was right on the surface of the sample, then if we moved the sample 2 mm toward the laser beam, the focus spot would be 2.9 mm behind the sample surface. When we fitted the third harmonic signal by  $C/w^4$ ,

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

From the Rayleigh range  $z_R$ , the focusing spot size was found to be 7.45  $\mu$ m when we used the 1" focal length lens.

#### 6. Self focusing.

The location and the dimension of the focus spot were measured by using the surface third harmonic signal was under low field condition. When we looked for optical breakdown threshold the field was a lot higher than that when we measured the surface third harmonic signal. The refractive index of fused silica is

$$\mathbf{n} = \mathbf{n}_0 + \mathbf{n}_2 \mathbf{I} \ . \tag{4}$$

Where  $n_0$ ,  $n_2$ , I are the linear refractive index, nonlinear refractive index, and irradiance of the laser beam, respectively. Thus the laser beam focused in fused silica was experienced by self focusing effect which moved the focus downstream and increased the maximum irradiance.

The correction factor for the laser maximum irradiance was a function of the laser power and the location  $z_0$  of the laser focus spot behind the sample surface. It was illustrated in figure 5.



Figure 5: Irradiance enhancement for different depths of focus.

When the location of the focus spot was 4 Rayleigh ranges or larger, then the irradiance correction factor followed the formula

$$\frac{I_{corr}}{I_0} = \frac{1}{1 - P/P_{sf}} .$$
(5)  

$$P_{sf} = \frac{3.73\lambda^2}{8\pi n n_2} .$$
(6)

(6)

Where

The nonlinear refractive index  $n_2$  is different for linearly and circularly polarized light, we have  $P_{sf}(lin) = 4.29$  MW, and  $P_{sf}(cir) = 5.89$  MW.

### 7. Stimulated Brillouin Scattering (SBS).

When we were looking for the optical breakdown threshold, we always wanted to be sure that we did not have Stimulated Brillouin Scattering because SBS reflected the pump beam back upstream and it was nearly phase conjugate to the pump beam, thus, it added with the pump beam very well. Then if SBS existed, the optical breakdown threshold would not be correct. We can estimate the SBS threshold for a focus Gaussian beam as follows:

$$Gain_{SBS} = g_0 I_0 z_R$$
  
=  $g_0 \frac{2P\pi w_0^2}{\pi w_0^2 \lambda}$   
=  $2g_0 \frac{P}{\lambda}$  (7)

Where  $g_0$  is the SBS gain factor, and it is a function of the material and  $\lambda$  was the laser wave length. The gain was about 30 at SBS threshold, then we could estimate the laser power at SBS threshold for CW light (1.064 µm) was 0.3 MW. The SBS build up time was 30 ns, then for a 10 ns, the SBS threshold was about 0.9 MW.

Experimentally, we found the SBS threshold in D1 fused silica for linearly and circularly polarized light to be 0.86 MW. Figure 6 showed the incident, transmitted, and amplified SBS beams. The SBS pulse propagated back into the laser get amplified and went back to the sample, it appeared about 50 ns after the main pulse, 50 ns was the round trip time of our experimental set-up.



Figure 6: Incident (red) and transmitted (blue) waveforms of linearly polarized light at the power slightly above the SBS threshold.  $w_0=17\mu m$ ,  $t_0(FWHM)=7.5ns$ .

The SBS threshold was nearly at a fixed laser power, then if we plotted the laser irradiance versus laser spot size we could find the SBS existed region as a function laser pulse durations. It was illustrated in figure 7.



Figure 7: SBS existed areas for two laser pulse durations, 7.5 ns and 30 ns.

## **III. Experimental results and discussion.**

# 1. Laser induced damage thresholds in fused silica for seeded and unseeded lasers.

After we put the beam waist on the surface of the sample, then we moved the focus spot 9.57 mm behind the surface of the sample, when we used the 1" focal length lens, the Rayleigh range of our laser beam were 238 µm in fused silica. Thus, we were sure that we did not generate surface damage on our samples when the focus spot was 9.57 mm behind the surface. After this we shot a single pulse at different spots in our sample and keep raising energy until optical breakdown happened. When we used a seeded laser (single longitudinal mode, 7.5 ns FWHM) then the energy difference between a 100% go and a 100% nogo was less than 2.0 %. This damage threshold was independent from different spots of a sample and different fused silica samples (A0, B0, B1, D0, D1, D3, D5) from Corning. We also used unseeded laser (multiple longitudinal modes), since every pulse of the unseeded laser was difference, we shot each spot in the sample 3000 shots, if we had a go in those 3000 shots then it was counted as a go, otherwise, it was a nogo, and this process repeated for ten times so we could have a probability of go and nogo at a certain pulse energy. The energy gap between a 100% go and a 100% nogo for the unseeded laser in this case was about 15%. The laser induced damage thresholds in fused silica for seeded and unseeded lasers were shown in figure 8.



Figure 8: Statistical and deterministic behaviors of optical breakdowns induced by an unseeded laser and a seeded laser.

By analyzing the traces of the incident and transmitted pump beams, the damage threshold power of A0 fused silica was  $374.2 \pm 4.0$  kW for linearly polarized light, and  $387.7 \pm 6.2$  kW at 1.064 µm. The damage threshold powers were different these two polarizations because the self focusing effect was different for them. The corrected irradiance followed the formula

$$I_{corr} = \frac{I_0}{1 - P/P_{sf}},\tag{8}$$

where the uncorrected irradiance was defined as,

$$I_{0} = \frac{2P}{\pi w_{0}^{2}}.$$
 (9)

The corrected threshold irradiance for linearly polarized was  $470.7 \pm 5.0 \text{ GW/cm}^2$ , and  $476.4 \pm 7.6 \text{ GW/cm}^2$  for circularly polarized light.

When we unseeded our laser, then it would run multimode, the gain bandwidth of YAG was about 30 GHz, the free spectral range of our laser was 250 MHz, however, not all of the cavity modes under the gain curve lased. If we assumed that there were 8 equally spaced modes under the gain curve lased, their phases were random, these modes interfered with each other, thus, the output pulse consisted of a lot spikes as shown in figure 9. The width of each spike was about 10 ps, approximately, and the power of the highest spike could be 4 times higher than the maximum power of the seeded pulse of the same energy.



Figure 9: The simulated temporal profile of unseeded and seeded laser pulses of the same energy.

This showed that the damage threshold average irradiance of unseeded laser beam could be four times smaller than that of the seeded laser beam. This strongly suggested that laser irradiance was the deciding factor for optical breakdown and the damage occurred on 10 ps time scale or shorter.

We measured the laser power at the start of the optical breakdown when the pulse energy was at and above the threshold pulse energy by inspecting the waveforms of the incident and transmitted beams. The waveforms of linearly polarized transmitted pulses at different energies were shown in figure 10.



Figure 10: Transmitted waveforms below and above optical breakdown threshold,  $w_0 = 7.45 \text{ mm}$ ,  $t_0 \text{ (FWHM)} = 7.5 \text{ ns}$ .

At 3.0 mJ pulse energy (fig. 10), it was slightly below the damage threshold then the laser beam transmitted through the fused silica sample undisrupted, the transmitted and the incident pulses were identical, and the sample remained undamaged if we shot more laser pulses at the same pulse energy into it. When we increased the pulse energy from 3.0 mJ to 3.05 mJ then we generated optical damage at every single shot. The optical breakdown started near the peak of the incident pulse. When we increased the pulse energy from 3.05 mJ to 5.27 mJ, the optical breakdown started at the laser power of about 372 kW on the rising edge of the incident pulse. These data showed that the optical breakdown in fused silica at this pulse duration (7.5 ns FWHM) started at the same laser power or at the same laser electric field strength independent of the incident pulse energy and also from the time measured from the start of the incident laser pulse to the beginning of the optical breakdown. Thus, we could see that this optical breakdown process was dominated by tunneling ionization.

#### 2. Damage morphology.

We also studied damage morphologies in the bulk of fused silica. The top and bottom views of the optical damages did not give us much information about the damage morphology. We also looked at the optical damage along the propagating direction of the pump beam. Figure 11 showed the optical damages in fused silica created by single longitudinal mode pulses whose energies were slightly above the damage threshold.



Figure 11: Optical damages in fused silica viewed along the propagating direction of the pump beam. The pump pulse energy was slightly above the damage threshold.

In figure 11, the dash line was the location of the pump focusing spot, the dot gave us an impression about the uncertainty  $(\pm 10 \ \mu\text{m})$  in the location of the focusing spot. The two curve lines gave us an impression about the focusing of the pump beam before the focusing spot and defocusing after that, and their length was about two Rayleigh ranges.. The optical breakdown started at the focusing point where the laser intensity was highest. Since the pump pulse energy was slightly above the damage threshold then the optical breakdown should start at the peak of the incident pulse. After the optical breakdown started, the pump beam was on the falling edge of the pulse, it was reflected, scattered, and absorbed by the plasma. In the optical breakdown process, we had two thresholds: a threshold to start the breakdown and a threshold to sustain it, and the threshold to start the breakdown was larger than the threshold to sustain it. The reflected beam and the incident beam were absorbed by the plasma, the tail of the damage was generated by this process until the laser intensity could not sustain the breakdown. The optical damage was very reproducible.

# 3. Focal size effect.

We measured damage threshold irradiances at radii of 7.5, 8.1, 12.7 and 17  $\mu$ m. The damage thresholds at 7.5 and 8.1  $\mu$ m fell below the SBS threshold, and they were equal within measurement error. However, the damage thresholds for radii 12.7 and 17  $\mu$ m lie above the SBS threshold. The optical breakdown process was probably assisted by SBS so the damage thresholds for these radii were slightly smaller.



Figure 12: Damage threshold irradiance at different beam radii.

The last data point on figure 12 is from a measurement that used a curved mirror to retroflect the laser beam back into the sample. This creates nodes and anti-nodes inside the sample. The threshold pulse energy was a little bit less than a third of that without the retroreflector. The indicated error bars for this data point are large because the alignment of retroreflected beam was imperfect. We conclude that the damage threshold is probably reduced by SBS, if present, but there is no apparent focal size effect on the damage irradiance.

#### 4. Surface damage.

We measured surface damage thresholds of D1 fused silica samples polished by an allalumina process at Alpine Research Optics. We did not use conventional cerium oxide polished silica samples because they have a low surface damage threshold apparently caused by the residual cerium oxide incorporated into the sample surfaces. In measuring the surface damage threshold, we first located the focal waist exactly at the input face of the sample by maximizing the surface third harmonic. We found a single-shot surface damage threshold by setting the pump pulse energy about 30% above the bulk damage threshold to ensure optical breakdown on every shot. The single-shot damage threshold was deduced from the irradiance at which transmission through the sample terminated. The single-shot damage threshold is statistical with an 18% standard deviation, and the highest damage threshold is 7% higher than the bulk damage threshold. The large standard deviation might be due to contaminants and defects on the sample's surface. To test this we conditioned the sample surface by raising the pulse energy slowly from 5% of the bulk damage threshold until breakdown occurs. The surface damage threshold measured this way is also statistical with an 11% standard deviation, and the highest threshold is also about 7% higher than the bulk threshold.

# 5. Influence of Yb<sup>3+</sup> doping on the damage threshold in fused silica.

We measured the bulk damage threshold of 1% Yb<sup>3+</sup>-doped fiber preform samples provided Liekki. However, the measurement was unreliable because the laser beam is distorted in core region due to optical index inhomogeniety of our samples. To avoid beam distortion we resorted to measureing the surface damage threshold. The measuring method was the same as outlined above. The highest surface damage threshold of the 1% Yb<sup>3+</sup>-doped fiber preform is equal within 2% to that of pure fused silica. This is a preliminary result, further study of the damage threshold of Yb<sup>3+</sup>-doped fused silica will be conducted.

#### 6. Electron avalanche model.

The electrons in the conduction band are generated predominantly by impact ionization from a small number of seed electrons that might be generated by multiphoton ionization. In the simplest electron avalanche rate model, the electron's density follows the rate equation [6].

$$\frac{dn}{dt} = \beta^{(8)}I^{8}(t) + \alpha nI(t) - \left(\frac{n}{\tau}\right)_{loss}.$$
(10)

where  $\alpha$  is the avalanche coefficient,  $\beta^{(8)}$  is the 8-photon absorption cross section because it takes 8 photons at 1.064 µm to excite an electron across the ban gap of fused silica,  $\tau$  is the free electron lifetime due to electron-hole recombination. Since the electron lifetime in fused silica is only 150 fs [7], optical breakdown is nearly instantaneous on the time scale of our 7.5 ns pulses. The electron avalanche model matched our optical breakdown thresholds provided  $\alpha \tau = 2 \ 10^{-4} \ \mu m^2/W$ .

## **IV. Conclusions.**

We have reported that the bulk damage threshold irradiance in fused silica was 471  $\pm$  5.0 GW/cm<sup>2</sup> and 476  $\pm$  7.6 GW/cm<sup>2</sup> for linearly and circularly polarized light, respectively, the pulse duration was 7.5 ns, and the radius of the focus spot was 7.45  $\mu$ m. The surface damage threshold is higher than the bulk damage threshold if the sample's surface is properly prepared. The surface damage threshold irradiance in 1% Yb<sup>3+</sup> doped fiber preform was equal that of pure fused silica.

The stimulated Brillouin threshold power in fused silica was  $851.0 \pm 2.9$  kW for linearly polarized light and  $852.0 \pm 2.3$  KW for circularly polarized light, for our pulse duration of 7.5 ns.

All of the reported damage threshold irradiances have been corrected for self focusing. Our measurements show that damage by ns pulses occurrs at a precise threshold irradiance, not a threshold fluence. The damage threshold irradiance was reduced by SBS, and there was no evident focal size effect. Optical breakdown occurred on a time scale of 30 ps or less.

We have also reported interesting and reproducible bulk damage morphology in fused silica.

## References

1. M. Soileau and M. Bass, IEEE J.Quan. Elec., QE-16, NO. 8, 814 (1980).

- 2. L. Gallais, J. Natoli, C. Amra, Optics Express, 10, 1465 (2002).
- 3. N. Bloembergen, IEEE J. Quan. Elec., QE-10, NO 3, 375 (1974).
- 4. T. Tsang, Phys. Rev. A, 52, 4116 (1995).
- 5. C. Carr, H. Radousky, A. Rubenchik, M. Feit, and S. Demos, Phys. Rew. Lett., **92**, 087401 (2004).
- 6. M. Lenzner, J. Kruger, S. Sartania, Z. Cheng, Ch. Spielmann, G. Mourou, W. Kautek, and F. Krausz, Phys. Rev. Lett., **80**, 4076 (1998).
- 7. P. Audebert et al., Phys. Rev. Lett. 73, 1990 (1994).

#### Acknowledgement:

Supported by Laboratory Directed Research and Development, Sandia National Laboratories, U.S. Department of Energy, under contract DE-AC04-94AL85000.