

The damage mechanism in borosilicate glass generated by nanosecond pulsed laser at 1.064 μm

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Abstract

We studied theoretically the laser-plasma interaction, and performed experiments to investigate the mechanisms giving rise to optical damage in Borosilicate glass using nanosecond laser pulses at wavelength 1064 nm. Our experimental result shows that the optical damage process generated by nanosecond laser pulses is the result of an optically induced plasma. The plasma is initiated when the laser irradiance frees electrons from the glass. Although it may be debated, the electrons are likely freed by multi-photon absorption and the number density grows via impact ionization. Later when the electron gas density reaches the critical density, the electron gas resonantly absorbs the laser beam through collective excitation since the laser frequency is equal to the plasma frequency. The laser energy absorbed through the collective excitation is much larger than the energy absorbed by multi-photon ionization and impact ionization.

Our experimental result also shows the plasma survives until the end of the laser pulse and the optical damage occurs after the laser pulse ceases. The plasma decay releases heat to the lattice. This heat causes the glass to be molten and soft. It is only as the glass cools and solidifies that stresses induced by this process cause the glass to fracture and damage.

We also show the experimental evidence of the change of the refractive index of the focusing region as the density of the electron gas changes from sub-critical to over-critical, and the reflection of the over-critical plasma. This reflection limits the electron gas density to be not much larger than the critical density.

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I. Introduction

Borosilicate glass (BK7) is an important optical material. The search for the correct optical damage mechanism in BK7 glass is important, since if we can identify the damage mechanism then we might find ways to increase the damage threshold and to make better optics.

We theoretically studied the laser-plasma interaction through a simple model of electron gas oscillating on a positive ionic background. This model shows that the electron gas strongly absorbs the laser beam when the plasma frequency of the electron gas is equal to the laser frequency. This absorption drives the electron gas density from sub-critical to over-critical and the refractive index at the laser frequency goes through an anomalous dispersion.

In the experiment, we have investigated the optical damage process in the bulk of borosilicate glass (BK7) created by nanosecond laser pulses. These experiments use tightly constrained parameters, including an extremely well behaved and characterized single frequency Gaussian laser beam focused tightly into the sample. Using tight focus and a single mode laser enables the investigation of fundamental damage thresholds and mechanisms without the complication of stimulated Brillouin scattering (SBS). In a high power laser application with large laser spot, the effect of SBS needs to be taken into account.

In this paper, we show experimental evidence that supports the idea that when the laser pulse starts to interact with the Borosilicate glass, a plasma forms, probably by the multi-photon absorption and impact ionization processes, and when this plasma density nears the critical density it strongly absorbs the laser energy through collective excitation, changing the material from highly transparent to totally absorbing within a fraction of a nanosecond. We also show experimental evidence of optical reflection from the plasma,

and optical lensing due to the plasma, which shows the plasma density changes from sub-critical to over-critical.

By studying the temporal profile of the broadband light emitted from the plasma, we show that the plasma survives longer than the laser pulse. Also, by performing a simple experiment we show that the physical damage such as cracks was formed after the end of the laser pulse, and this result is consistent with the calculated temperature of the absorbed volume.

We will describe a simple model of laser plasma interaction in part II, the statement of the problem in part III, our experimental set-up in part IV, a description of our laser beam in part V, and we will discuss our experimental results on the reflection and absorption of the plasma in part VI. We will show experimental evidence of the change in plasma density from sub-critical to over-critical in part VII, and evidence of when the physical damage formed in part VIII. We will show how long the plasma survives in part IX; we propose a possible optical damage mechanism in borosilicate glass in part X, and conclusions in part XI.

II. A simple model of laser plasma interaction

Under the influence of an external driving laser field, the free electron can oscillates on a positive ionic background [Ref. 1]. Schematically, we can describe the motion of a single electron in the ensemble of this electron gas in the following figure,

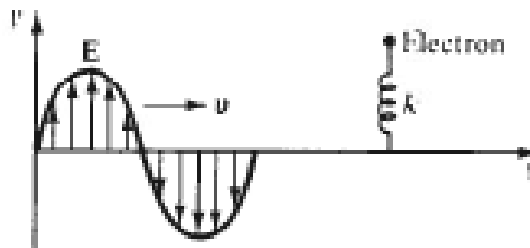


Figure 1: The motion of an electron in the ensemble of electron gas on a positive ionic background.

The equation of motion of this electron is as follows:

$$m \frac{d^2 y}{dt^2} = F_{binding} + F_{damping} + F_{driving}. \quad (1)$$

The driving force comes from the external laser field, the binding and damping forces come from positive ionic background and the electron gas. Eq. (1) can be written as follows:

$$m \frac{d^2 y}{dt^2} = m\omega_0^2 y + m\gamma \frac{dy}{dt} - qE_0 e^{-i\omega t}, \quad (2)$$

where ω_0 is the natural oscillating frequency and γ is the damping constant, m is the electron mass, and $-q$ is the electron charge. The above equation can be solved for the displacement $y(t)$ in the following form:

$$y(t) = \frac{q}{m[(\omega_0^2 - \omega^2) - i\gamma\omega]} E_0 e^{-i\omega t}. \quad (3)$$

The resulting polarization is

$$P = Nfqy(t) = \frac{Nq^2}{m} \frac{fE_0 e^{-i\omega t}}{(\omega_0^2 - \omega^2) - i\gamma\omega}. \quad (4)$$

where N is the molecular density and f is the fraction of these molecules which have electrons oscillate at the frequency ω_0 . The polarization can also be written as

$$P = \varepsilon_0 \chi E_0 e^{-i\omega t}, \quad (5)$$

where χ is the susceptibility. The dielectric constant is

$$\varepsilon = \varepsilon_0 (1 + \chi) \quad (6)$$

$$\varepsilon = \varepsilon_0 \left[1 + \frac{Nq^2}{m\varepsilon_0} \frac{f}{(\omega_0^2 - \omega^2) - i\gamma\omega} \right]. \quad (7)$$

The wave vector of the laser field in this medium is

$$k = \omega \sqrt{\varepsilon\mu_0}. \quad (8)$$

From the wave vector we can find the change in the refractive index seen by the laser field and the absorption coefficient

$$\Delta n_f \approx \frac{Nq^2}{2m\epsilon_0} \frac{f(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2}, \quad (9)$$

$$\alpha \approx \frac{Nq^2\omega^2}{m\epsilon_0 c} \frac{f\gamma}{(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2}. \quad (10)$$

These formulae for Δn_f and α are functions of the natural oscillating frequency of the electron gas with respect to the positive background.

Let us assume that the electron gas is displaced from the positive ionic background a distance Δx as described in Fig. 2 [Ref.2].

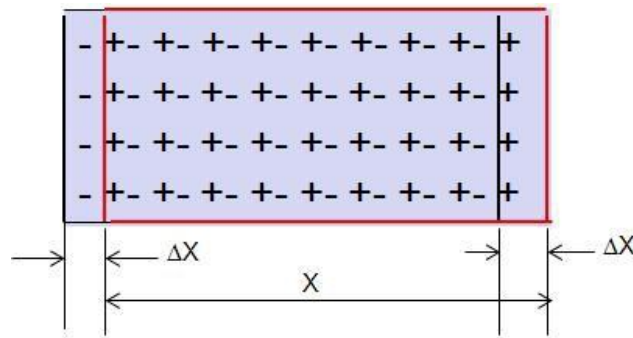


Figure 2: The electron gas is displaced a distance Δx with respect to the positive ionic background and it is pulled back to the equilibrium.

The displacement of the electron gas with respect to the positive ionic background generates an electric field of the form,

$$E = \frac{n\Delta X e}{\epsilon\epsilon_0}. \quad (11)$$

This electric field generates a recovery force acting on each electron, the equation of motion of each individual electron is

$$-\frac{ne^2\Delta X}{\epsilon\epsilon_0} = m \frac{d^2\Delta X}{dt^2}. \quad (12)$$

The above equation of motion describes the oscillation of the electron gas; it shows that the natural oscillation frequency of the electron gas is the plasma frequency,

$$\omega_0 = \sqrt{\frac{ne^2}{\epsilon\epsilon_0 m}}. \quad (13)$$

Figure 3 shows the change of the refractive index seen by the electric field of the laser and the plasma absorption coefficient shown in Equations (9) and (10), where the plasma frequency have been normalized by the laser frequency.

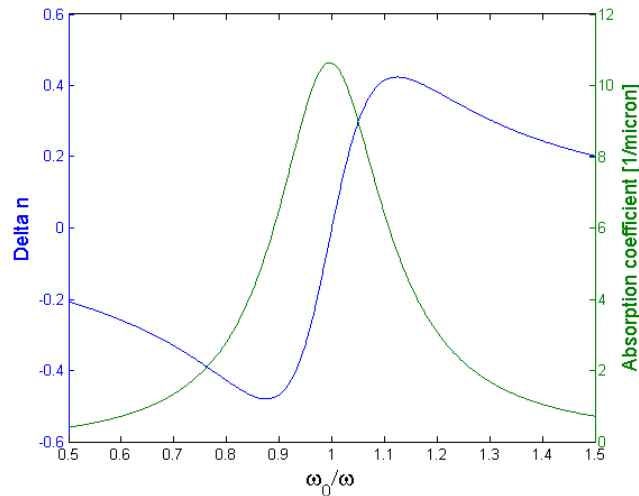


Figure 3: The change in the refractive index seen by the electric field of the laser and the plasma absorption coefficient.

When the free electron density is equal to the critical density then the plasma frequency is equal to the laser frequency. The plasma resonantly absorbs the laser light through the collective excitation. This absorption combined with impact ionization increases the free electron density past critical meaning the plasma frequency is greater than the laser frequency and the plasma absorption is reduced. The plasma becomes opaque to the laser light, and the light is strongly reflected.

Whether the plasma is sub-critical or over-critical can be distinguished by the weak lensing created by the plasma. When the free electron density is sub-critical, its refractive index is decreased so the plasma forms a weak diverging lens (i.e. negative lens). When the free electron density is over-critical, the opposite is true, the refractive index is increased and the plasma forms a weak converging lens (i.e. positive lens). In Sec. VII, we will discuss how the sign of the plasma induced lensing can be detected by measuring changes in the beam profile after it has been transmitted through the plasma.

III. Statement of the problem

There are several experimentally observable phenomena which can be inferred from the above model of laser plasma interaction. When the pulse energy of the laser exceeds the damage threshold, then the laser pulse will be transmitted through the BK7 window until there is an abrupt stop to the transmission. However, this stop in transmission is evidence of a plasma and not evidence of immediate damage, which damage actually occurs 100's of nanoseconds later. The evidence that a critical plasma exists when the transmission of the laser transmission stops includes:

1. The laser beam is strongly absorbed by the plasma after the transmission of the laser beam through the sample was stopped rather than being scattered by optical damage such as cracks.
2. The transition of the plasma from subcritical to over-critical can be observed by the lensing effect of the plasma on the transmitted beam. A subcritical plasma creates a negative focal length lens, and an over critical plasma creates a positive focal length lens.
3. There is no scatter from cracks until after the laser pulse.
4. The plasma persists even after the end of the laser pulse as evidenced by broadband white light generation.

IV. Experimental set-up

A diagram of the experimental setup is shown in Figure 4. We used a single-longitudinal-mode, injection-seeded, Q-switched YAG laser operating at 1.064 μm . The temporally smooth pulse is repeatable with a pulse-to-pulse amplitude variation of less than 1.5 %. In order to extract a single laser pulse or a set of N pulses from our 10-Hz laser while keeping the laser oscillator locked to the seed laser we used a beam shutter synchronized to the Q-switch signal. The shutter opened 40 msec before the first pulse and closed at 40 msec after the last pulse. The pulse energy was varied using a half wave plate and a high-energy cube polarizer. We used a spatial filter with a 200- μm -diameter wire die at the focus of an one-to-one imaging telescope followed by an adjustable iris to make the laser beam a close approximation to a TEM_{00} profile. We also used four fast phototubes (Hamamatsu, R1193U-01) to record the temporal profiles of the incident, full transmitted,

observed transmitted, and reflected pump beams, and a photomultiplier (Hamamatsu, 1P28) to record the broadband light emitted from the optical breakdown region. A HeNe probe beam co-aligned with the high power beam was displayed on a screen positioned downstream from the sample. The sample was mounted on a motorized 3-dimensional translation stage, and we focused the pump beam into the sample by a 1-inch focal length, best form lens manufactured by CVI.

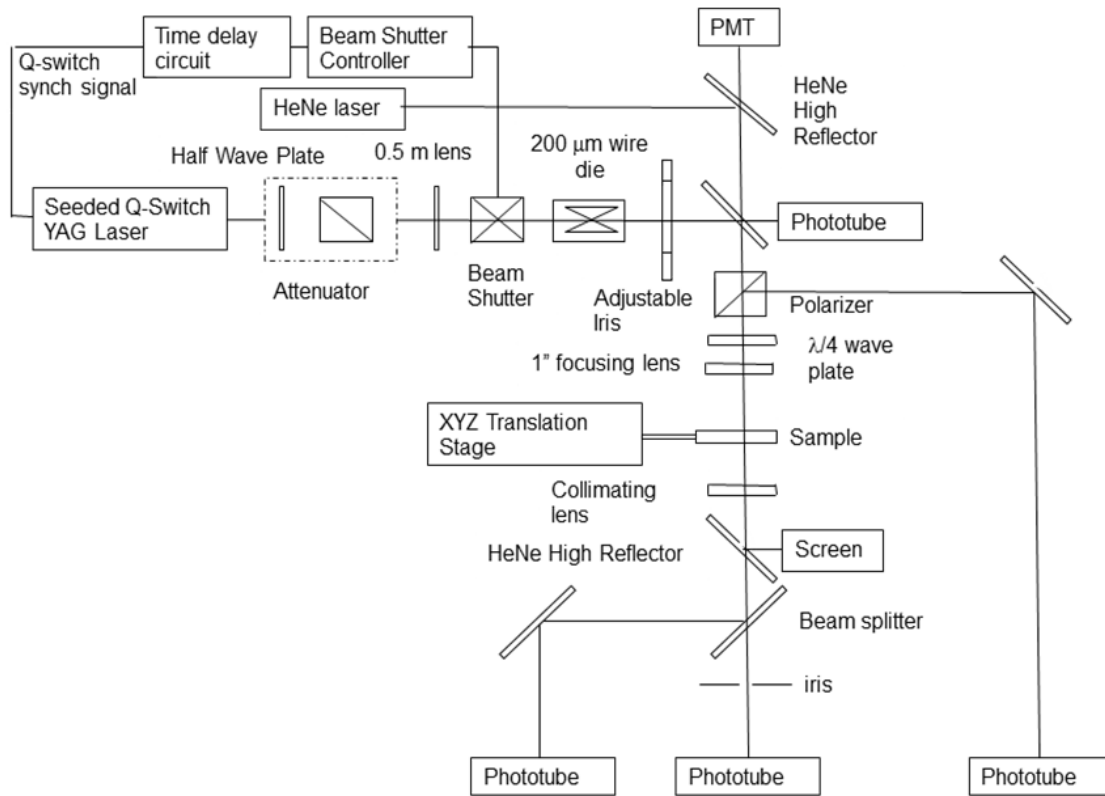


Figure 4: Experimental set-up

V. Spatial and temporal profiles of laser pulses

Va. Spatial beam profile measurement

Figure 5 shows the spatial profile of the beam before the focusing lens. The spatial profile is close to the TEM₀₀ mode, ensuring that it focuses to a nearly diffraction-limited spot size without hot spots.

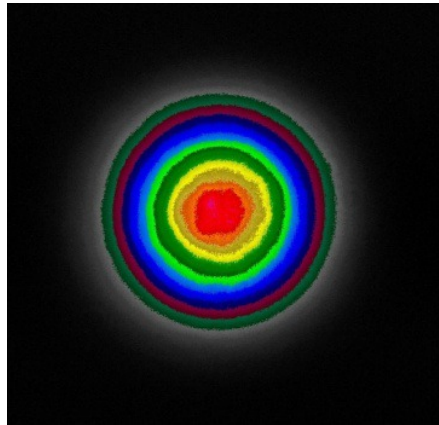


Figure 5: The spatial profile of our laser beam before the focusing lens.

The laser beam is focused to a spot diameter of $15\ \mu\text{m}$ at a location $3\ \text{mm}$ below the sample surface. The size of the focus spot and its location were precisely measured and set by using the surface third harmonic signal [Ref. 3]. Since the focus is small, the laser power at breakdown is below the SBS threshold, eliminating the complications of SBS.

V b. Temporal profile of laser pulses

The temporal profile of the high power $1064\ \text{nm}$ pulse is shown in Fig. 6.

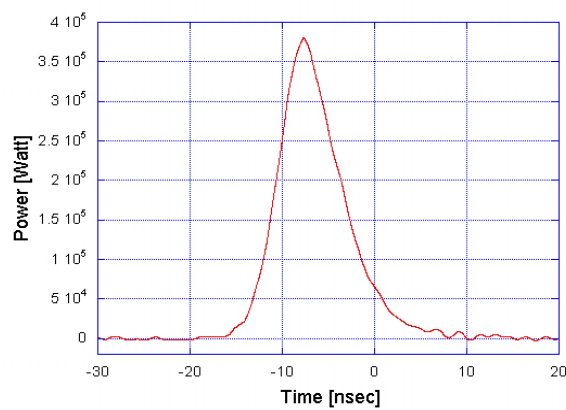


Figure6: The temporal profile of our laser pulse. The FWHM is 8nsec.

The pulsed laser which was used in this experiment is single longitudinal mode, with a full width at half maximum of 8 ns, and a pulse energy variation from pulse to pulse of approximately $\pm 1.5\%$.

VI. When the laser beam is reflected and strongly absorbed by the plasma

Figure 7 shows representative experimental results for optical damage in BK7. The key feature to notice is the sharp cut-off in transmitted light immediately after the plasma is created.

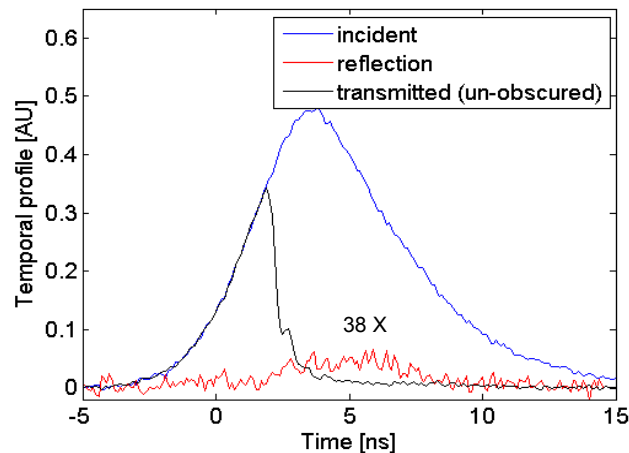


Figure 7: Plasma reflects the laser beam after the transmission of the laser beam through the borosilicate window was stopped.

When the transmission of the laser beam through the borosilicate window is stopped, the plasma reflects a small fraction of the laser beam back along the beam path. This scattered laser energy is estimated to be less than 3% of the incident pulse energy after accounting for a collection angle of 0.2 steradians and assuming a cosine scattering. After the transmission of the laser pulse was stopped, the laser pulse is almost entirely absorbed by the plasma at the focus spot.

The plasma strongly absorbs the laser beam driving the electron gas density above the critical value. The plasma absorption is decreased as plasma frequency exceeds the laser frequency because in this regime the plasma is opaque to the laser. The subcritical plasma

in front of the focus strongly absorbs the incident and reflected laser beams, this effect makes the plasma grows backward toward the laser source.

The experimental result in Fig. 7 shows that the borosilicate glass changes from highly transparent to nearly total absorbing the laser beam within a fraction of a nanosecond because of the build-up of critical plasma at and in front of the focus spot.

VII. Evidence of sub-critical and over-critical plasma

A subcritical plasma, with its negative change in refractive index, creates a negative focal length lens defocusing the light. In contrast, an over-critical plasma, with its positive change in refractive index, creates a positive focal length lens focusing the light. Thus, by comparing the signals from the obscured and un-obscured apertures we can distinguish whether the plasma was subcritical, critical, or over-critical. The obscured aperture has a donut pattern with light being blocked in the center and transmitted around the periphery. A positive lens focuses more light to the center where it is blocked for the obscured aperture, and a negative lens directs more of the light around the obscuration. The signals were balanced for the obscured and unobscured apertures such that they are equal in the absence of plasma lensing. The experimental result on the evidence of sub-critical and over-critical plasma is shown in Fig. 8 (a) and (b).

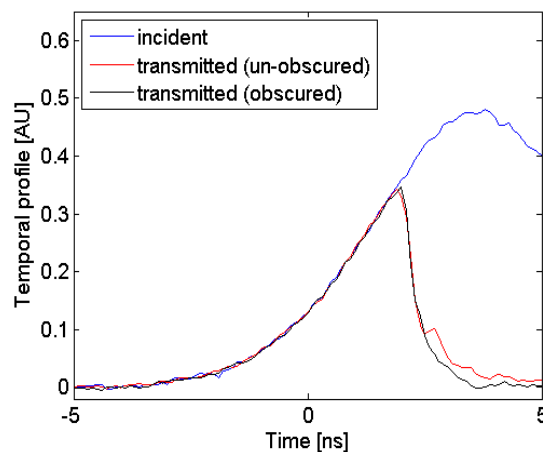


Figure 8(a): The enhancement and decrease of the obscured transmitted laser beam with respect to the fully transmitted one.

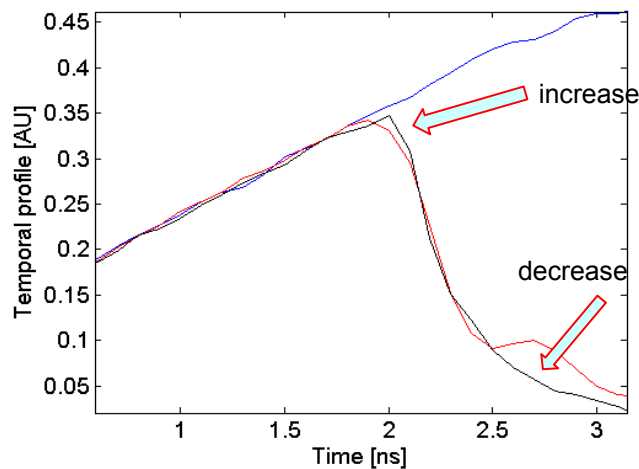


Figure 8(b): Zoom into the locations of the enhancement and decrease of the obscured transmitted laser beam.

Figure 8 shows that at 2 ns the obscured signal is greater, i.e. there is a sub-critical plasma. From about 2.2 ns to about 2.4 ns the obscured and unobscured signals are roughly equal and the attenuation increases rapidly. Thus, on average the plasma has become critical and reached its peak absorption. The volume of the plasma is also growing during this time but has not yet started moving upstream along the incoming laser path. At 2.5 ns the bulk of the plasma transitions to over-critical and the absorption coefficient decreases corresponding to a bump in the transmitted signal. The positive lensing evidenced by the unobscured signal being larger than the obscured signal also indicates an over-critical plasma.

VIII. When does the physical damage occur?

The generation of a plasma inside the glass is not necessarily sufficient to create damage. For example, we have observed similar plasma generation in BK7 using femto-second laser pulses without fractures and optical damage. The mechanism for optical damage (by optical damage we mean cracks and scattering inclusions) is when stress inside the material exceeds the tensile strength of the material. There are two reasons indicating optical damage does not happen until long after (100's of nanosecond or longer) the damage inducing laser pulse.

The first reason for thinking that happens long after the laser pulse is that we do not observe any scattering during the damage pulse. As shown in Figure 7, almost all of the light incident on the plasma is absorbed, with less than 3% of the light scattered. In contrast the scattered light from cracks surrounding the plasma region would be much higher than 3%.

To investigate how much light is scattered from optical damage, we created a damaged spot using the first laser pulse. After the damage was formed we used a second pulse at the same spot to measure the reflected signal. The scattered signal from the second pulse was very high until optical breakdown at the damage site results in a plasma which engulfs the damage and absorbs all of the light. Figure 9 shows the reflected/scattered signal for this experiment with the region of interest occurring before -2 ns when the plasma enveloped and destroyed the scattering. In order to see the scattering signal without the 4% Fresnel reflection from the front surface, we subtract the blue curve from the red curve as shown in Figure 10.

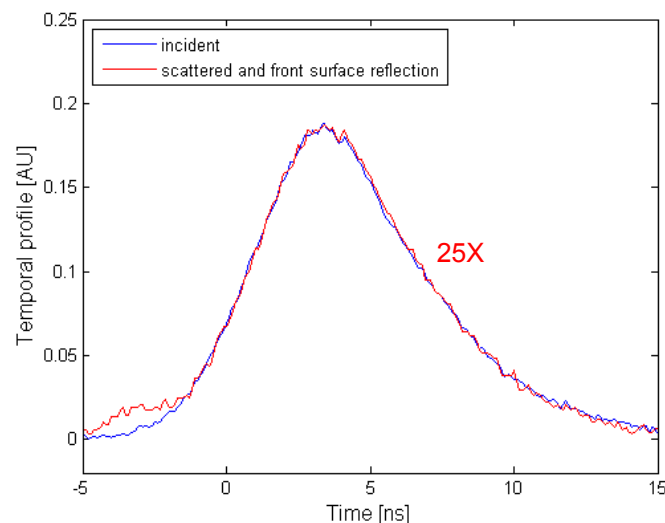


Figure 9: The temporal profile of the scattered light from the damage and the reflected light from the front surface is compared with the temporal profile of the incident pulse.

A direct comparison of the scattering signals in Figure 10 and Figure 7 is complicated by the fact scattering from optical damage in Figure 10 only occurs during the very tail of

the rising edge. However, in Figure 10 both the red and blue curves have the same shape up until -4 ns, and this region can be used to estimate the magnitude of scattering from cracks and optical damage. It is plainly evident that scattering due to optical damage is orders of magnitude larger than the scattering from the plasma observed in Figure 7. As shown in Figure 11, the cracks created by optical damage radiate away from the plasma region in all directions. If the optical damage had occurred during laser pulse in Figure 7, we should expect to see scattering similar to what is observed on the leading edge of the pulse in Figure 10. Clearly, there is no evidence of cracks in the scattering signal shown in Figure 7. The comparison between the two figures 10 and 7 shows that the physical damage such as cracks was not formed until the end of the laser pulse.

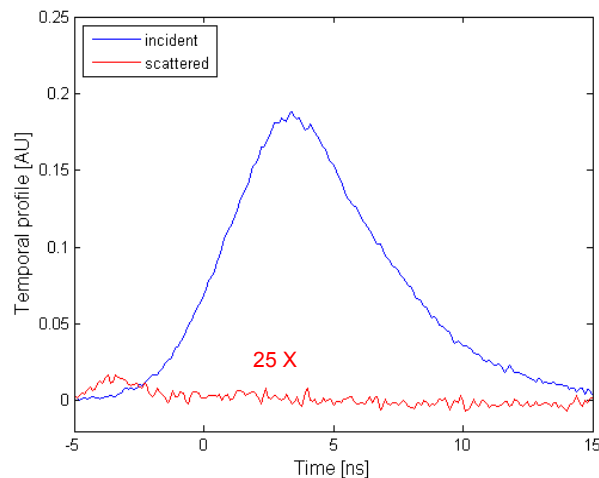


Figure 10: The temporal profile of the scattered light from the damage generated by the first laser pulse.

The absence of crack and optical damage during the laser pulse should not be surprising if we consider the physics of the super-heated glass in the plasma region. A simple back-of-the-envelope calculation should elucidate this physics. For simplicity, we assume that pump beam induces a plasma in the Borosilicate window at the peak of the pulse. From this point to the end of the laser pulse is 5 ns. The pulse energy is 3.3 mJ. The focus spot diameter is 15 μm , and the Rayleigh range is 250 μm . From the damage morphology

shown in figure 11[Ref. 4], we see that a half of the laser pulse energy was deposited into the volume from the focus back one Rayleigh range to the laser source.

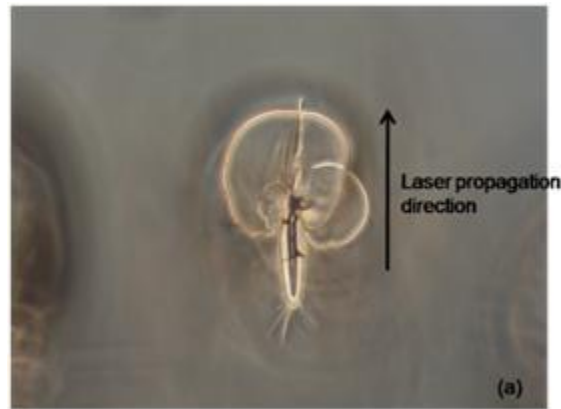


Figure 11: The damage morphology in Borosilicate glass, this damage is generated by a laser pulse slightly greater than the damage threshold.

Taking into account the convergence of the laser beam toward the focus, this volume is about 1.5 times the cylindrical volume whose diameter is 15 μm and the length of a Rayleigh range. This volume is equal to 6.63×10^{-8} cc.

The heat diffusion length is of the following form

$$l^2 = \frac{kt}{C_p \rho} \quad (14)$$

where k is the thermal conductivity, C_p is the specific heat, ρ is the density of Borosilicate glass, and t is 5 ns. The heat diffusion length is only 50 nm at 20°C , and at the Borosilicate glass melting temperature of 715°C , the heat diffusion length may be two or three times larger than that at 20°C , it is still much smaller than the dimension of the absorbed volume. Thus, the heat generated from the absorption of a half of the laser pulse in the above volume stays in this volume. We estimate that at the end of the laser pulse, the peak temperature is in the range of 20000°K . At this temperature, the material is molten and soft; no cracks can form in soft material.

IX. How long does the plasma persist?

It is only after the plasma dies down and the glass begins to cool that we expect damage to occur. Thus, it is important to determine how long the plasma persists after the laser pulse. The broadband light emitted by the plasma indicates how long the plasma lasts. This broadband light was collected by a PMT and the result is shown in fig. 12.

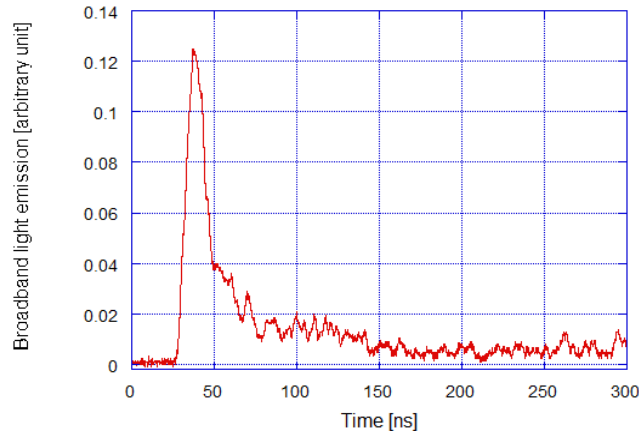


Figure 12: Temporal profile of the broadband light emitted from borosilicate glass.

The broadband light was emitted right after the transmission of the pump beam through the borosilicate stopped. This broadband light consists of two parts, a spike and a long tail. The width of the spike is about 20 ns, and a long tail (persisting longer than 100 ns). Thus, the plasma survives much longer than the end of the 8 ns laser pulse.

The possible reason for the long decay time is the plasma was not only induced optically, it was also induced thermally. The temperature of the absorbed region was calculated in the range of 20000 ⁰K, the band gap of borosilicate glass is 4.28 eV [Ref. 5], and the number of molecules per cubic centimeter (cc) is about 3.3×10^{22} . The thermally induced equilibrium plasma density can be estimated to be

$$3.3 \times 10^{22} \exp\left(-\frac{E_g}{kT}\right) = 3 \times 10^{22} \exp\left(-\frac{4.28}{1.73}\right) = 2.79 \times 10^{21}/\text{cc}$$

The critical density at 1.064 μm is $2.22 \times 10^{21}/\text{cc}$ [note: the free electron mass was used in the derivation].

Figure 12 shows that the plasma survives for more than 20 ns, and it was shown the plasma can be sustained for several minutes by a burst of laser pulses from high repetition rate laser [Ref. 6].

XI. The proposed damage mechanism of optical damage in borosilicate glass generated by nanosecond pulsed laser at 1.064 μm

In the section VI, we have shown the critical plasma exists when the transmission of the laser beam through the sample was stopped, and the critical plasma strongly absorbed the laser beam. In the section VII, we showed experimental evidence that the free electron density changes from sub-critical to over-critical, and in section VIII, the physical damage such as cracks were not formed at the laser focal spot until after the end of the laser pulse. Finally, in section IX we showed that the plasma persists long after the end of the laser pulse.

From the above experimental results, we propose a damage mechanism of optical damage in borosilicate glass generated by nanosecond pulsed laser at 1.064 μm :

1. A sub-critical plasma is initiated by some mechanism, most likely multi-photon absorption and impact ionization.
2. The near-critical and critical plasma resonantly absorbs the laser beam through collective excitation. This absorbed energy is much larger than the energy absorbed through the multi-photon absorption and impact ionization; it is responsible for the damage morphology.
3. When the plasma is over-critical, it reflects the laser beam. This reflection limits the electron gas density.
4. The plasma grows backward toward the laser source.
5. The plasma survives much longer than the laser pulse. As the plasma decays it releases energy into the lattice in the form of heat, and this thermal energy heats up a region starting at the laser focus and extending along the oncoming laser path.

6. The heated region cools down induces stress. When the stress exceeds the tensile strength of the borosilicate glass, it forms cracks and damages the sample.

XI. Conclusion

We have shown that the critical plasma exists when the transmission of the laser beam through the sample is stopped, and the critical plasma strongly absorbs the laser beam through collective excitation. We also showed experimental evidence that the free electron density changes from sub-critical to over-critical, and physical damage such as cracks do not form at the focal spot until after the end of the laser pulse. We also showed evidence that the critical plasma still exists to the end of the laser pulse.

From this experimental evidence, we propose a damage mechanism of optical damage in borosilicate glass generated by nanosecond pulsed laser at 1.064 μm . A plasma is initiated by some mechanism involving the absorption of laser energy. Most likely the mechanism for initiating the plasma is multi-photon absorption and impact ionization. The main mechanism for transferring energy from the laser pulse to the plasma is the collective excitation of the critical plasma. At the end of the laser pulse, this absorbed laser energy raises the absorbing volume to very high temperature, approximately 20,000 K. Thus, the material is molten, and cracks cannot form in the molten or soft material. The cracks were formed when the heated region cools and its volume changes creating stress that exceed the tensile strength of the now solid glass. Cracks are formed when the stress becomes greater than the tensile strength of the material.

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Questions and Answers

Q. In the graph that you have up there, which is the plasma reflection for a Drude dielectric, the sharpness of that transition and how much reflection you have at the critical density depends upon the collision time. So in your calculation here, what did you take as the collision time and where did it come from.

A. Oh, I don't do any calculation on the collision time. Let's say that in a nanosecond there isn't much reflection. I think my speculation because of the spot size of about 15 micrometers is in the range of nanoseconds. The spot size in the femtosecond regime is about 1 micrometer so the Rayleigh branch we are hopping for is about two or three times smaller than in the other case. The reflected beam had less subcritical plasma to go through. That's why we see the reflection at the beginning, but after that, after about 20 or 30 pulses, the plasma region grows too big and the light goes in and is absorbed by the subcritical plasma. And, when you reflect it, it is absorbed again and that's why we don't see it. After 20 or 30 pulses, that is my speculation. Did I answer your question?

Q. Yes. I was just making a comment. If the lifetime is very short as it is in a solid, you can go to densities that are higher than critical and still not get very much reflection.

A. How come. Why would it go too high above the critical density? If it goes to 1.1, 1.2, 1.3 above critical, then it would go opaque. So, the laser beam is not absorbed, it is reflected out.

Q. It depends upon the sharpness of the curve you have drawn in your upper graph. That depends on the laser frequency and lifetime...

A. Which one?

Q. The one you just had up. The dielectric constant as a function of... Yes. That one. The plasma frequency, the same graph as a function of density. The sharpness of that transition depends upon the plasma and materials parameters.

A. Oh, you are talking about the width of this curve, the absorption. Yes. Thank you.