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Time-resolved imaging of material response during laser-induced bulk damage in SiO2

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ABSTRACT

We report on time resolved imaging of the dynamic events taking place during laser-induced damage in the bulk of fused silica samples with nanosecond temporal resolution and one micron spatial resolution. These events include: shock/pressure wave formation and propagation, transient absorption, crack propagation and formation of residual stress fields. The work has been performed using a time-resolved microscope system that utilizes a probe pulse to acquire images at delay times covering the entire timeline of a damage event. Image information is enhanced using polarized illumination and simultaneously recording the two orthogonal polarization image components. For the case of fused silica, an electronic excitation is first observed accompanied by the onset of a pressure wave generation and propagation. Cracks are seen to form early in the process and reach their final size at about 25 ns into the damage event. In addition, changes that in part are attributed to transient absorption in the modified material are observed for delays up to about 200 microseconds.

Keywords: Laser damage, laser-induced modifications, damage mechanisms, fused silica

1. INTRODUCTION

Laser-induced damage of optical components is a fundamental applied science problem that represents the key performance limiting factor in lasers designed to deliver high energy and/or high power pulses. Under operational conditions similar to those found in Inertial Confinement Fusion (ICF) class laser systems, damage is initiated at pre-existing defects that, through mechanisms that may be different for different materials, lead to strong coupling of the laser energy into localized regions of the material.[1,2] This can result in irreversible material modifications that interfere with the propagation of subsequent laser pulses, causing beam energy loss and downstream intensity modulation that may lead to additional damage. Because of the time and cost to repair or replace optics, the operational budget of such laser systems strongly depends on the amount of laser-induced damage.

The material science and laser physics involved in a damage event can be separated into five distinct phases:
1) The incorporation of the defect in the material during manufacturing or preprocessing steps
2) The response of the precursor (via its electronic structure and physical characteristics) to the incident laser light
3) The response of the host material to localized energy deposition, leading to formation of a damage site
4) Modification of the electronic and structural properties of the host material within a damage site, both during initiation and each subsequent shot
5) The interaction of the modified host material with subsequent laser pulses that result in damage growth

Our hypothesis for the general description of the damage timeline is given in Fig. 1. It should be emphasized that this description is a hypothesis that needs confirmation or denial. At the present time, a relatively small amount of data is available to accurately describe the events within the timeline. Yet our search for precursor identification and theoretical models for damage behavior are often based on the implicit assumption that this hypothesis is correct. Because the current generation of ICF-class laser systems is designed to operate very close to the damage threshold of their optical components, it is imperative that we achieve a fundamental understanding of all stages involved in the damage process. This understanding is expected to expand our options for managing damage initiation and growth in the optical components during operation of such laser systems.

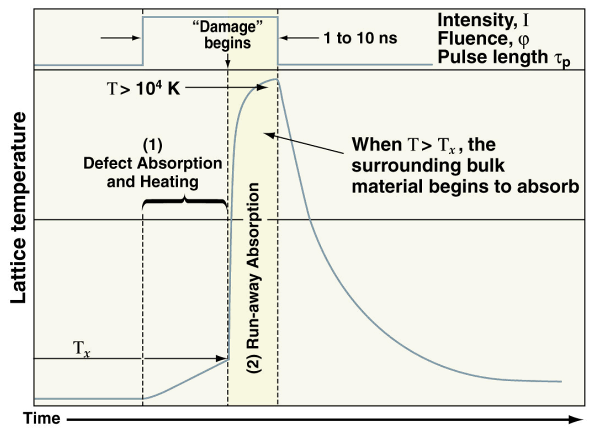

Fig. 1. The current hypothesis on the timeline of events involved during laser induced damage (M. L. Spaeth, unpublished)

Past research has focused almost exclusively on phase 1[2,3], phase 2[4-9] and phase 4[10-16] although the ultimate characteristics of the damage site are governed by processes in phases 3 and 5 for damage initiation and growth, respectively. In spite of advancements in understanding, the exact mechanisms and the timeline of these processes remain largely unknown. It is the objective of this research to significantly broaden understanding of these areas.

Post-mortem examination of laser-induced damage sites demonstrates material modifications associated with changes in material structure at the atomic level[11,12,14] accompanied by mechanical damage.[13,14] The formation of a pressure wave following damage initiation in bulk of KDP material has been demonstrated but the spatial resolution was limited.[17] More recently, we reported on the temporal behavior of the emission during a bulk damage event within optically transparent materials.[18] It was observed to have a blackbody character that reached temperatures on the order of 10^4 K. We conclude from these studies that the changes of the material structure can be attributed to the combination of the *high temperatures and pressures occurring during a laser damage event.*

Our research and other reports in the literature provide numerous descriptions of experimental approaches that can be used to probe different time-bites within a damage event. Before the temperature of the precursor reaches T_x (seen in the timeline), there is work by various groups that demonstrate imaging of transient electronic excitations that do not lead to an observable permanent change in the material.[19,20] These measurements have been performed using excitation pulses on the order of 100 fs while the images were based on shadowgraphy or detection of fluorescence. Russo et al. have performed a considerable amount of work to detect and image the shockwaves propagating on both sides of the interface during front surface ablation in fused silica using ps and ns pulses.[21-23] These experiments were performed using shadowgraphy; several interpretations of their results have been proposed. Using the same experimental technique, Russo's group was able to image ejected material during ablation of silicon.[24-26] Although they demonstrated that this process is delayed by at least a few hundreds of ns, they were not able to resolve the kinetic properties of the ejecta. Still, their work suggests that shadowgraphic microscopy could help us resolve both the propagation of the shock wave and the kinetic properties of the ejecta. By building on their experience, our approach is to use a similar experimental configuration that can provide better spatial resolution and more than one time-delayed image per damage event. In addition, our ability to access the most advanced modeling tools will help with the interpretation of experimental observations.

This work represents a first step in an effort to elucidate the processes involved during a damage event in optical materials. Specifically, we demonstrate a time resolved microscopy system capable of imaging the dynamic material changes during a damage event throughout the entire timeline. The capabilities of this instrumentation were explored by studying intrinsic bulk damage in fused silica. We demonstrate that, although the system is not yet fully optimized, the underlying mechanisms of a damage event can be recorded with high spatial and temporal resolution. Such experimental results will enhance our understanding of the damage process as well as guide the development of detailed models.

2. EXPERIMENTAL SETUP

In order to study the dynamic behavior of the material following damage initiation, we have developed a microscopy system capable of providing time resolved images using a pump-and-probe configuration. We expected that the timeline of events involved in laser induced damage (up until the damage site takes its final form) extends to ~100 µsec or longer from the beginning of the laser pulse. Optical delays are not feasible for such long delay times. This task required the use of separate pump and probe lasers synchronized by external triggering to achieve time delays covering this entire timeline as depicted in Fig. 2a. In this preliminary work, we have used Q-switched Nd:YAG lasers, with the pump laser operating at 355 nm with 3 ns pulse duration (FWHM) and the probe laser operating at 532 nm with 4.5 ns pulse duration.

The imaging system has been designed to perform polarization sensitive shadowgraphic microscopy. The schematic layout in Fig. 2a indicates the use of the probe laser for polarized, pulsed illumination of the damage site (created by the preceding pump pulse). The damage site and the surrounding volume are imaged with high spatial resolution (on the order of 1 µm) using a lens system composed of a 5X long-working distance objective followed by a second set of lenses which effectively provides an additional 5X magnification. The image is split using a polarizing beam splitter cube into two orthogonal polarization components and recorded by separate CCD detectors. The parallel polarization image is used to monitor changes in transmission due to changes in the material's complex index of refraction. The orthogonal polarization image is used to map stress via depolarization and can image pressure waves generated after the initial energy deposition and residual stress fields.

Figures 2b and 2c show the parallel and perpendicular polarization images, respectively, of the same bulk damage site in fused silica (final form) as recorded by our experimental system. The left side in the image of the damage site exhibits a network of cracks. In the right hand side of the image of the damage site, an elongated region that has been modified by the pump pulse is observed but is largely free of cracks. Due to the angle between the direction of propagation of the pump pulse with respect to the image plane, not all of the modified region created by the pump can be maintained in focus by the imaging system (with a depth of focus of ~ 50 microns). For this reason, we chose to keep the right hand side of the image out of focus in order to better capture the processes taking place in the region where the cracks are forming. Thus, the parallel image shows in focus the cracks and the "core region" of the damage site. This image is complemented by the perpendicular polarization image component (in Fig. 2c) which reveals the stress fields causing depolarization of the probe light. It must be noted that only one transient set of the two polarization image components can be captured at any pre-set pump and probe delay time from each damage event. However, the use of a focused pump beam (deterministic damage via intrinsic damage initiation) enables one to study the processes throughout the entire timeline by capturing images of different, but similarly evolving, damage events.

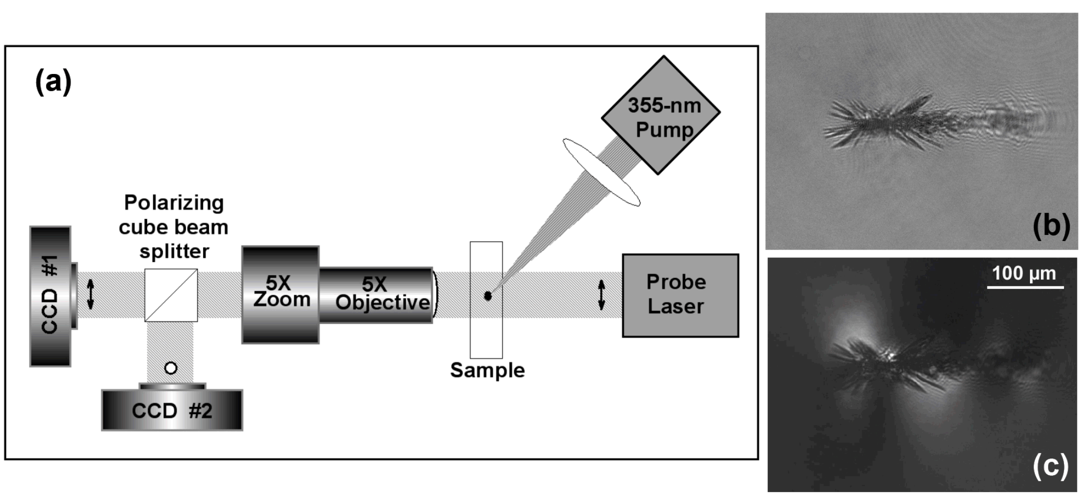

Fig. 2. a) Experimental layout of time-resolved, polarization-sensitive microscopic imaging system in the trans-illumination geometry used to study the evolution of bulk damage. Typical images of the same bulk damage site (final form) in fused silica showing b) the parallel and c) the perpendicular polarization image components captured using our experimental system. In all images, the pump pulse enters from the left hand side.

3. EXPERIMENTAL RESULTS

Figure 3 shows the transient images captured during bulk damage events using pump and probe delay times between 0 and 25 ns. Three images are shown for each time delay: the parallel polarization transient image (left), the perpendicular polarization transient image (center), and the parallel polarization final image (right) of the same damage event. This time window represents the interval in which the pump pulse is illuminating the material and the energy deposition takes place (captured in the image at 0-ns delay) and the subsequent interval that finishes when the cracks and other major morphological features of the final damage site have formed. For bulk damage in fused silica using our 3-ns pump laser, the experimental results indicate (by comparing the transient and final images) that cracks reach their final size within the first 25-30 ns.

The transient images shown in Figure 3 indicate that, following the energy deposition, a pressure wave is launched (also referred to in the literature as a shock wave) and is best viewed in the perpendicular polarization images (bright regions). The location of energy deposition appears absorbing in the parallel polarization image (dark regions indicate transmission losses of the illuminating probe pulse) and may be related to absorption by the plasma formed. However, the parallel polarization images indicate that the affected region remains absorbing after the termination of the pump pulse, most probably due to subsequent material modification. The cracks appear to start forming with the onset of energy deposition and launch of the pressure wave.

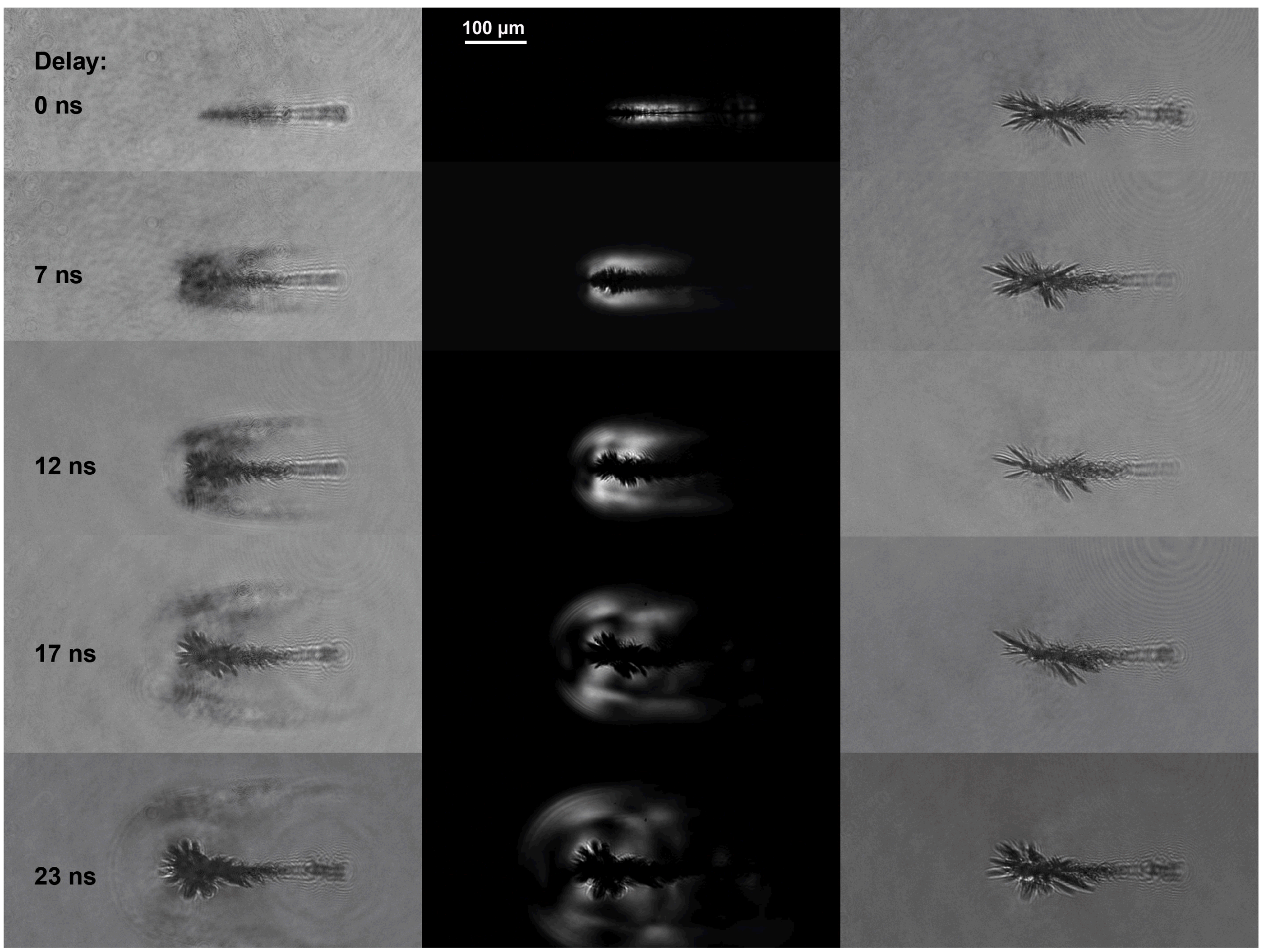

Fig. 3. Transient images captured at different delay times during a damage event. Three images are shown for each time delay: the parallel polarization transient image (left); the perpendicular polarization transient image (center); the parallel polarization final image (right) captured at the end of the same damage event.

Figure 4 shows transient images captured in the 30 ns to 120 ns time window. The main features in these images are associated with the propagation of various pressure waves. The pressure wave (p-wave) launched at the onset of the damage process propagates outwards while additional stress waves follow behind. Among these, the most prominent is what we believe to be the s-wave and appears in the perpendicular polarization image as a dark front propagating at a lower speed than the bright front of the p-wave. It is expected that the stress field involved in the s-wave would not cause any depolarization of the probe light in our system. These images indicate that there are other stress waves emitted at later times, as can be seen in the 113 ns delay time image after the initial pressure wave has propagated outside the imaged area. We will further investigate this effect in a future publication.

Comparison of the parallel polarization images at each delay time shown in Figure 4 (transient and final) indicates that although the main morphological features are practically identical, their contrast is higher in the transient image. To better capture this effect, we calculated the ratio image via pixel-by-pixel division of the transient over the final image. Results are shown in Fig. 5 for delays between 50 ns and 500 µsec. We postulate that the difference in the intensity and the subsequent formation of “bright” features in the ratio image arises from the presence of transient absorption associated with material modification. Specifically, the cracks are visible in the ratio images up to about 300 ns delay. This may suggest that the transient absorption is associated with transient defects formed on and/or near the crack surface immediately after its formation. The results suggest that, within the first 300 ns, this transient defect population decays possibly via lattice rearrangement. The ratio images also show the “core” region of the damage site as a bright feature indicating that it is also giving rise to a transient absorption. However, this region remains absorbing for a much longer time up to about 150 µsec delay time. We postulate that this “core” region is where the material has melted. This latter transient absorption may then be assigned to the host material properties in a transient state, i.e. a liquid phase at high temperatures. Thus far, we have not determined (by experiments or modeling) the temperature above which the material exhibits this absorption. We intend to address this issue in the future.

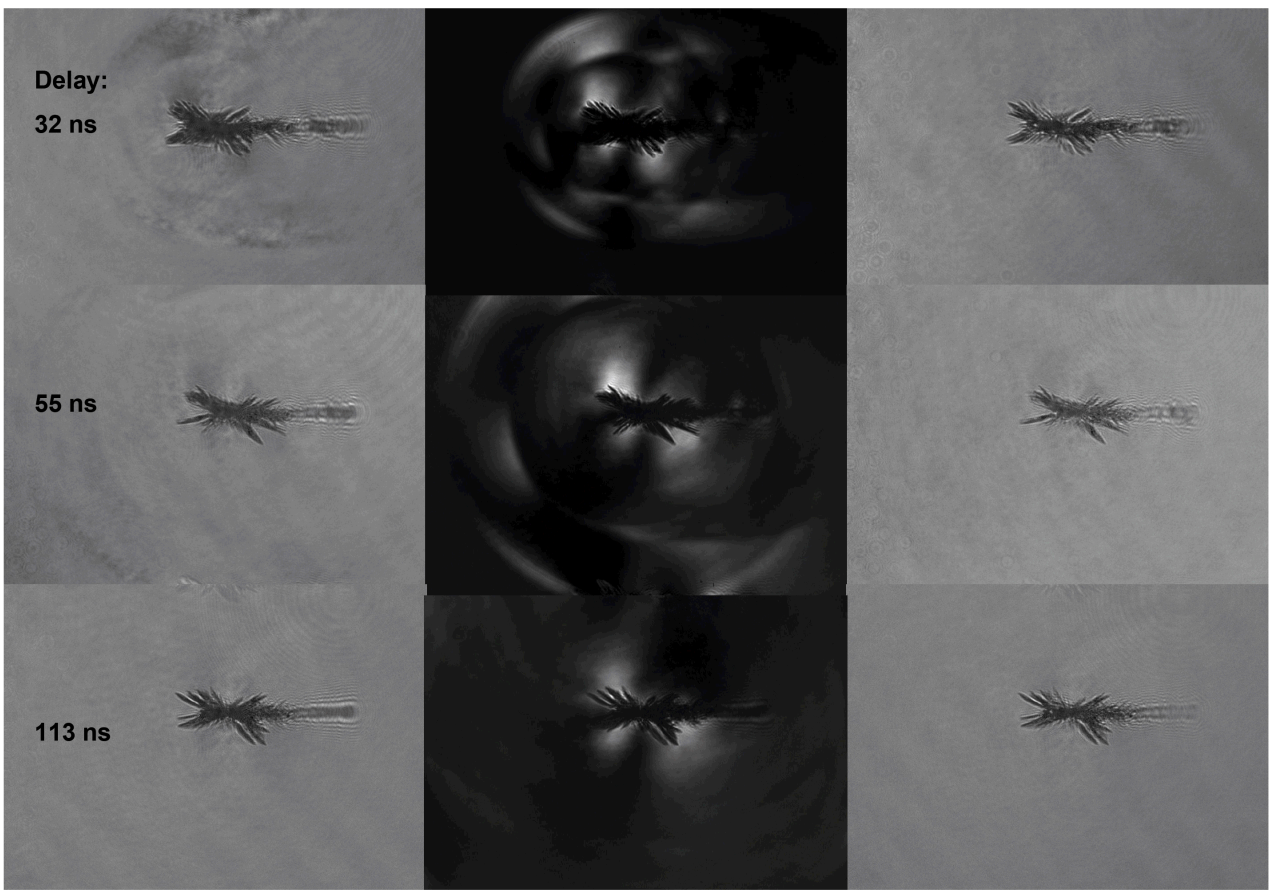

Fig. 4. Transient images captured at delay times between 30 and 120 ns showing the evolution and propagation of various stress waves. The morphology of the damage sites during this time window is not changing significantly.

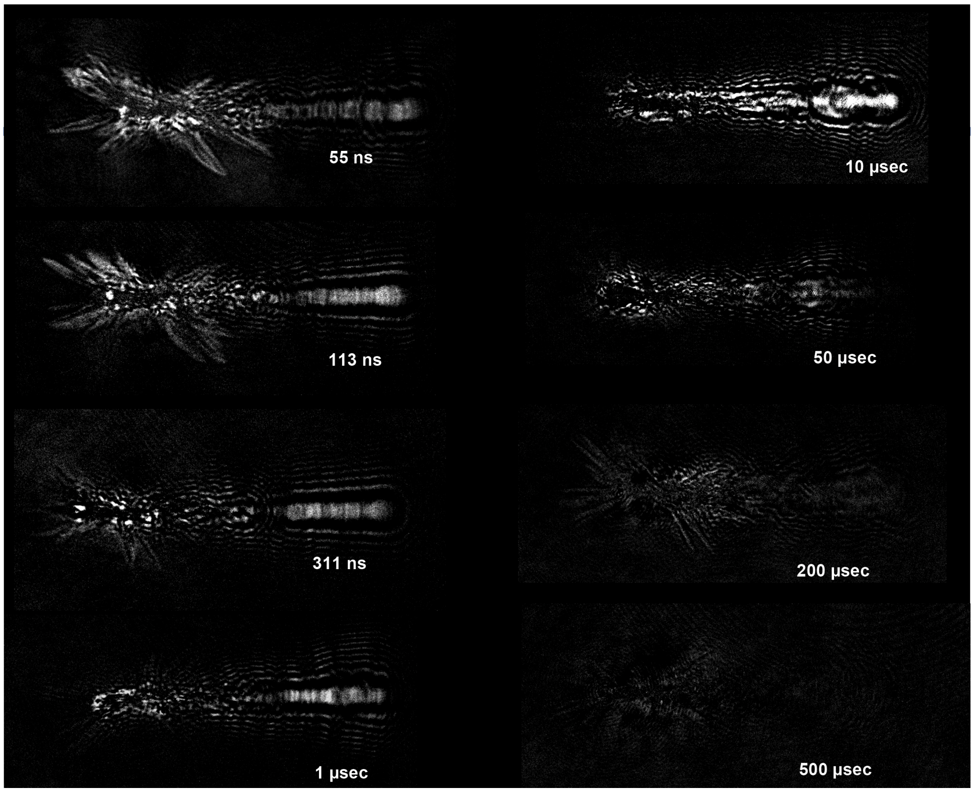

Fig. 5. Ratio images (transient divided by final parallel polarization image components) for delay times between 50 ns and 500 µsec showing features associated with the presence of transient absorption. The cracks are visible during the first 300 ns while the "core" (melted) region exhibits transient absorption for delays up to about 150 µsec.

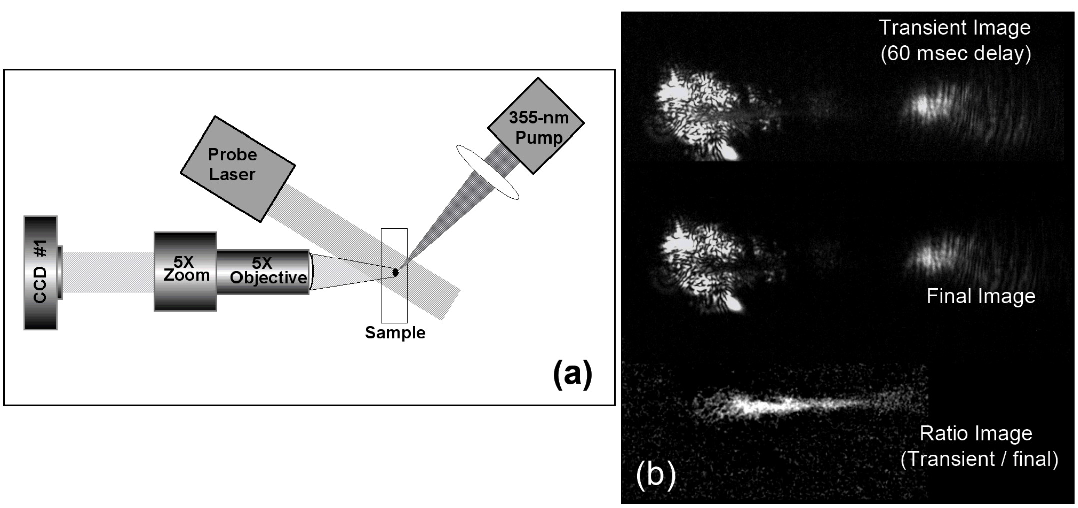

Fig. 6. a) Experimental layout of time resolved microscopic system in the backscattering geometry. b) Transient image at 60 msec delay (top) and the corresponding image at the end of the damage process (middle). The ratio image (bottom) indicates the presence of a 7 µm in diameter volume that has not reached its final state at this delay.

The energy deposition by the pump (damage inducing) laser pulse leads to melting of a localized region of the material. The return of this melted material to its solid phase may be considered as the last step of the damage process. We hypothesize that this phase change will lead to changes in the scattering properties of the material that may be captured by our imaging system if properly configured. To test this concept, we changed the orientation of the probe beam to a backscattering geometry as shown in Fig. 6a. This setup allowed us to capture backscattering images of the damage sites, for both transient and final stages. Figure 6d shows the transient image at 60 msec delay and the corresponding final image. The image quality is low due to interference effects from scattering of the coherent laser light at multiple scattering sites within the damage site. However, these two images are not identical. This difference is better highlighted in the ratio image shown in Figure 6b (bottom). There is an elongated volume of about 7 μm in diameter where the ratio image indicates the material is still in a transient phase at this delay time. This effect terminates at about 70 msec delay. We postulate that the origin of this effect may be due to scattering of the probe laser light on micro-voids formed by volume contraction during cooling of the material in liquid phase. Since this effect terminates at about 70 msec delay, we assume that this indicates the return of the material to its final state (solid phase).

The time resolved microscopic images of intrinsic bulk damage in fused silica shown in this work provide a preliminary evaluation of dynamic parameters of the damage process. Specifically, by monitoring the propagation of the initial pressure wave (p-wave), we can estimate its speed of propagation which was found to be about 5.7 Km/sec (which is the speed of sound in fused silica). Similarly, the speed of propagation of the s-wave was found to be about 3.4 Km/sec. In addition, the speed of propagation of the cracks during the first about 20 ns is approximately 2.5 Km/sec with the speed thereafter declining as cracks stop growing at about 25-30 ns delay. These experimental results are summarized in Fig. 7.

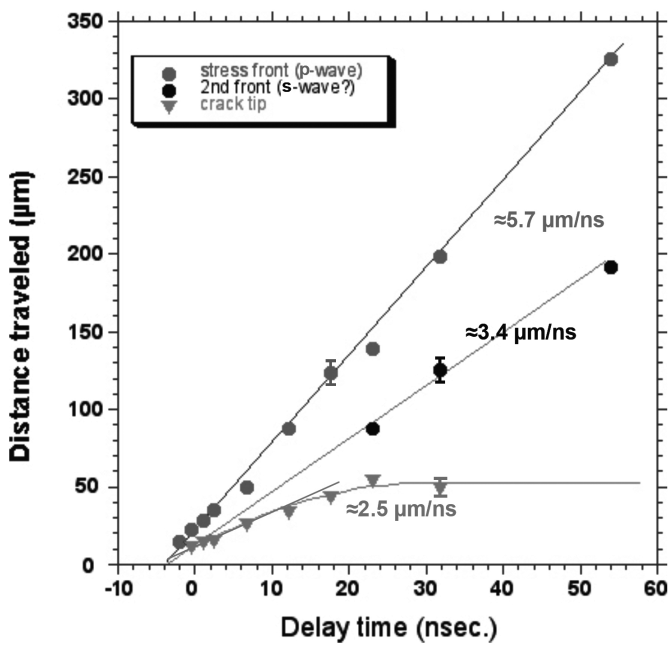

Fig. 7. Preliminary evaluation of dynamic parameters of the damage process using the time resolved images including the propagation of the p-wave, the s- wave and the crack tips.

4. DISCUSSION

The time resolved images shown in this work revealed information on the processes involved during bulk damage in fused silica through the entire timeline. The results suggest: a) The initial region of absorption is on the 1 μm scale but the damage "core region" expands to ~30 μm in the 5-25 ns time window; b) The cracks grow during the first 25 ns at a speed of ~2.5 Km/sec; c) The crack regions are absorbing after formation up until ~300 ns delay; d) The core (melted) region remains absorbing until ~200 μs delay; e) Additional stress waves are emitted after the initial p-wave; f) the melted region (7 μm in diameter) returns to solid phase at ~70 ms delay.

Although the results shown in Figs 3-6 demonstrate that the experimental system is capable of imaging the damage evolution with high spatial and temporal resolution, the relatively long pulse duration of the probe beam introduces a major limitation associated with the spatial resolution of the dynamic features such as propagating stress waves, growing cracks and expanding plasma. Specifically, the temporal width of the probe pulse (~5 ns) causes the spatial resolution of the propagating p-wave and cracks to be on the order of 30 μm and 12 μm, respectively (pulse duration x speed). Since the spatial resolution of the static images is on the order of 1 μm, a probe laser with pulse duration shorter than ~150 ps will be able to optimize the spatial resolution of the dynamic image features. This will allow the spatial resolution of the dynamic events to be similar to that of the static image (about 1-2 μm). This will help resolve the spatial width of the emitted stress fields which, in turn, will increase the sensitivity of the system by increasing the image contrast (the propagating stress fields will be confined to a smaller image area). This upgrade on the probe laser is underway and future reports will be based on results using this optimized system.

We anticipate that the upgraded system will also help us better resolve the energy deposition process by acquiring images during the pump (damage-inducing) laser pulse with adequate spatial and temporal resolution. We hope to be able to resolve the physical dimensions of the initial electronic excitation region leading to plasma formation and resolve its dimensions as it expands as well as determine the transmission loss through it (absorption coefficient). This will enable the employment of hydrodynamic codes to further enhance our understanding of the early fundamental processes. We also hope to resolve the width of the s- and p- waves as well as additional emitted stress waves. Overall, we believe such experiments will help develop a detailed understanding of the energy dissipation mechanisms and dynamic material response.

The time evolution of the transient absorption by the cracks and the core region (as shown in Figure 5) is in agreement with the transient absorption observed using pump-and-probe damage testing.[27] This agreement provides an explanation on the mechanisms behind these previous findings but also substantiates the argument that the origin of the results shown in Figure 5 is the presence of enhanced absorption (as opposed to scattering).

Understanding the processes involved during a damage event can help better manage damage in large aperture laser systems. The size of the damage sites depends on a) the total energy absorbed which is directly related to the energy coupling mechanisms and b) on the response of the material to the localized energy deposition. Furthermore, damage growth is determined by the same process as well as by additional mechanism(s) that lead to the coupling of the laser energy at a pre-existing damage site. Future work will focus on these areas. However, the work presented here benchmarks the experimental approach and provides information on fundamental material response behaviors that are independent of the damage initiation conditions.

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