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# Pump and probe damage testing for investigation of transient material modifications associated with laser damage in optical materials

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## ABSTRACT

Laser-induced breakdown in the bulk of transparent dielectric materials is associated with the generation of extreme localized conditions of temperatures and pressures. In this work, we perform pump and probe damage testing experiments to investigate the evolution of transient absorption by the host material arising from modifications following confined laser energy deposition in fused silica and DKDP materials. Specifically, we measure the size of the damage sites observed in the region of spatial overlap between the pump and probe pulses versus probe time delay and energy. Results of this proof-of-principle experimental work confirm that material modifications under extreme conditions created during a damage event include transient optical absorption. In addition, we found that the relaxation times of the induced absorption are very distinct for DKDP and SiO<sub>2</sub> even under identical excitation conditions, on the order of 100 ns and 100  $\mu$ s, respectively.

**Keywords:** Laser-induced material modifications, optical materials, localized energy deposition

## 1. INTRODUCTION

In large bandgap materials, laser intensities in excess of  $10^{11}$  W/cm<sup>2</sup> are required to achieve intrinsic breakdown.<sup>1</sup> However, defects in the material can initiate localized breakdown at more than two orders of magnitude lower intensities. Laser damage caused by nanosecond scale pulses in practical optical materials is nearly always due to precursors, i.e. very small absorbers not present in the ideal material. Even though the amount of energy absorbed by such precursors can be as small as tens of nanojoules, the energy density is high enough that the precursor is vaporized and forms a plasma with temperature in the eV range ( $10^4$  K). The hot high pressure ( $\sim 30$  GPa) plasma generation launches a shockwave into the surrounding material.<sup>2,3</sup> This localized energy is fast dissipating and is accompanied by a sequence of transient material modifications that lead to the formation of a void.<sup>3,4</sup> Material modifications associated with laser damage arise from key fundamental processes that are not well understood, such as solid state material response to localized extreme conditions, energy transport mechanisms through complex material phases, and material displacement and lattice transformation. Moreover, the sequence of events remains largely unknown.

In this work, we present initial results using pump and probe damage testing to investigate the localized dynamics of transient optical absorption of the modified material following laser-induced breakdown. We explore the dependence of the transient absorption on size of initial energy deposition volume (from pump pulse) and probe laser fluence. We also estimate the absorption relaxation times for bulk DKDP and bulk fused silica, chosen for their very different structural, mechanical and thermal properties.

## 2. EXPERIMENT

Pump-probe measurements can be used to obtain information on complex transient phenomena. The general principle is the following. A sample is exposed to a first (pump) pulse, which generates some kind of excitation (or other modification) in the sample. After an adjustable time interval, a second (probe) pulse is used to irradiate the sample, and through its interaction with the material (as manifested using a variety of techniques) obtain information regarding transient changes introduced by the pump pulse. By monitoring the probe signal

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as a function of the time delay, one obtains information on the decay of the generated excitation, or on other processes initiated by the pump. The temporal resolution is fundamentally limited only by the pulse duration. In particular for the pump-probe laser-induced studies proposed in this work, the probe beam can also be absorbed and thus change the final (time equal infinity) state.

In this work, the pump-probe technique is integrated within a novel damage testing system that allows detection of damage events in the bulk of optical materials with very high spatial resolution.<sup>5</sup> Moreover, the delay between pump and probe pulses can be varied over a wide range, from nanoseconds to microseconds, and even milliseconds, in contrast to those achieved by optical delay lines. This approach can be realized by using two independent, nanosecond Q-switched Nd:YAG laser systems that are synchronized within five nanoseconds using an electronic pulse delay generator. A sketch of the experimental setup is shown in Fig. 1. The pump pulse is obtained from a first Nd:YAG, Q-switched laser with output at the fundamental (1064 nm) and 2nd and 3rd harmonics [532- and 355-nm with  $\sim 3$ -ns Full-Width-Half-Maximum (FWHM)]. The pump beam is used to induce damage sites in the bulk of the material and is kept at a fixed fluence just above the damage threshold of the material (with 10% shot-to-shot variation). The probe beam is obtained from a second Nd:YAG, Q-switched laser with output at the fundamental (1064 nm) and 2nd and 3rd harmonics (with  $\sim 7$ -ns FWHM at 355-nm) which is aligned to spatially overlap the pump beam inside the sample, with comparable or larger spot size than that of the pump. A mechanical, computer controlled fast shutter (FS) enables single pulse operation. The time delay, pulse widths and energies of pump and probe pulses are monitored in real time using fast photodiodes (PhD). A He-Ne laser beam is focused by a 250-mm focal length cylindrical lens (CL) through the back of the samples to illuminate the tested volume and detect bulk damage events (see Fig. 2). A CCD camera coupled to a microscope equipped with a long-working-distance objective lens is used to record high-resolution light-scattering images ( $2 \mu\text{m}$  per pixel) in order to monitor changes in the morphology of bulk damage sites induced by the pump pulse in the presence of the probe background fluence. The materials investigated here were conventional grown DKDP and UV grade fused silica. Both samples were  $5 \times 5 \times 1 \text{ cm}^3$  plates with optical quality finish on four sides. Figures 2(a) and 2(b) illustrate in more detail the pump-probe damage testing configurations and beam paths within the sample for two different excitation methods used in this study, spontaneous and intrinsic bulk damage init

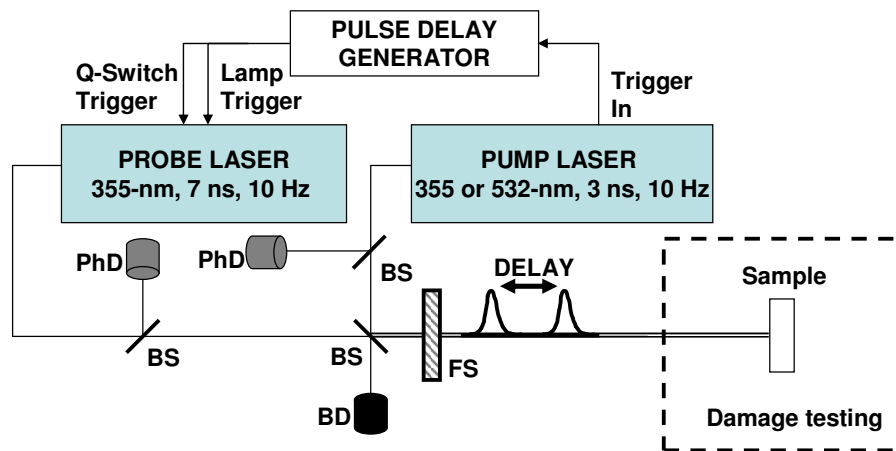


Figure 1. Schematic view of pump and probe damage testing using two independent, nanosecond Q-switched Nd:YAG laser systems (BS-beam splitter, PhD-photodiode, BD-beam dump, FS-fast shutter).

For the case of spontaneous damage in DKDP [see Fig. 2(a)], the pump and probe beams at 355-nm are co-propagating and focused within the bulk of the material using a 200-mm cylindrical lens (i.e., slit beams at the sample location). We note that no spontaneous bulk damage is observed in fused silica. The pump peak fluence was fixed at  $9 \text{ J/cm}^2$  (gaussian beam profile with 2.2 mm by  $60 \mu\text{m}$ ) and resulted in a damage pinpoint density of  $\sim 400 \text{ mm}^{-3}$  (see Ref. 5). The probe fluence was varied and kept below  $17 \text{ J/cm}^2$  to minimize the amount of damage due to probe pulse alone (gaussian beam profile with 2.4 mm by  $110 \mu\text{m}$ ). The individual damage sites created separately by the pump and probe pulses were  $5 \mu\text{m}$  and  $10 \mu\text{m}$  in size, respectively. In

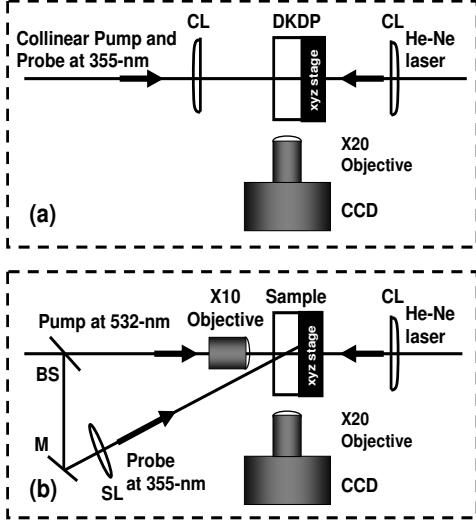


Figure 2. Closeup view of beam paths to the sample (dotted region in Fig. 1) in (a) collinear, one-color and (b) non-collinear, two-color pump and probe geometries for spontaneous and intrinsic damage, respectively (CL-cylindrical lens, SL-spherical lens, M-mirror).

this experimental configuration, the spatial distribution of pump and probe fluences along the slit beams leads to a distribution of sizes of damage sites within the interaction region, as illustrated by the light-scattering image shown in Fig. 3(a) (with  $5 \mu\text{m}$  per pixel resolution). We have assumed that the largest observed damage sites correspond to the spatial overlap of pump and probe pulses at peak fluences and thus their sizes will be reported as a function of probe peak fluence.

In order to achieve intrinsic bulk damage in DKDP and  $\text{SiO}_2$ , intensities in excess of  $200 \text{ GW}/\text{cm}^2$  at 532-nm (for 3-ns pulses) need to be employed. For this purpose and to prevent surface damage on the substrates, we have used a  $10\times$  objective in the pump beam path, as shown schematically in Fig. 2(b). This two-color, non-collinear pump-probe geometry allows for independent control of the probe beam size at the interaction region (i.e., focused using a separate spherical lens, SL). Shot-to-shot deterministic damage within the bulk can be achieved above the breakdown threshold. Therefore, the pump intensity at 532-nm was fixed at  $\sim 440 \text{ GW}/\text{cm}^2$  for both DKDP and  $\text{SiO}_2$ . The probe fluence at 355-nm (7-ns) was varied up to  $80 \text{ J}/\text{cm}^2$  in silica and up to  $28 \text{ J}/\text{cm}^2$  in DKDP. We note that, under identical excitation conditions, the damage sites in DKDP are  $\sim 4\times$  larger than those observed in  $\text{SiO}_2$ , as illustrated in Fig. 3(b). In addition, typical damage sites in fused silica consist of two pits with diameters of  $\sim 40 \mu\text{m}$  and  $\sim 20 \mu\text{m}$ , respectively, with the latter always upstream from the focus.<sup>6</sup>

### 3. RESULTS AND DISCUSSION

With the above described experimental configurations, we monitored the size of the damage sites observed in the pump and probe interaction region as a function of probe time delay at fixed probe fluences. Specifically, the overall size of damage sites is evaluated from light-scattering images in terms of cross-sectional area of the damage region, i.e. width  $\times$  height. Figure 3(c) illustrates the procedure for measuring the size of the damage area and comparison of damage sites from pump only to those observed after pump and probe interaction in  $\text{SiO}_2$ .

The results of pump and probe damage testing in DKDP are presented in Figs. 4(a) and 4(b) for spontaneous and intrinsic bulk damage, respectively. The size of the damage area in the pump and probe interaction region was measured as a function of probe time delay from 1 ns up to 500 ns for fixed probe fluences (not all shown). Similarly, measurements of the damage area for the case of intrinsic bulk damage in  $\text{SiO}_2$  are summarized

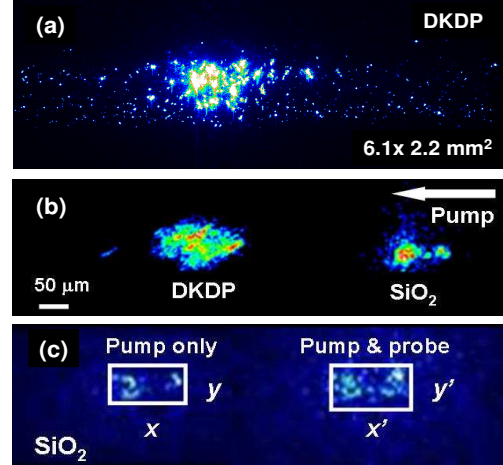


Figure 3. Typical scattered light images of bulk damage sites in the pump and probe interaction region for (a) spontaneous damage in DKDP and (b) intrinsic damage in DKDP and  $\text{SiO}_2$  under identical excitation conditions. (c) Size assessment of the damage region is performed in terms of cross-sectional area (i.e.,  $x$ - $y$ ) of damage sites (example shows damage sites in  $\text{SiO}_2$ ).

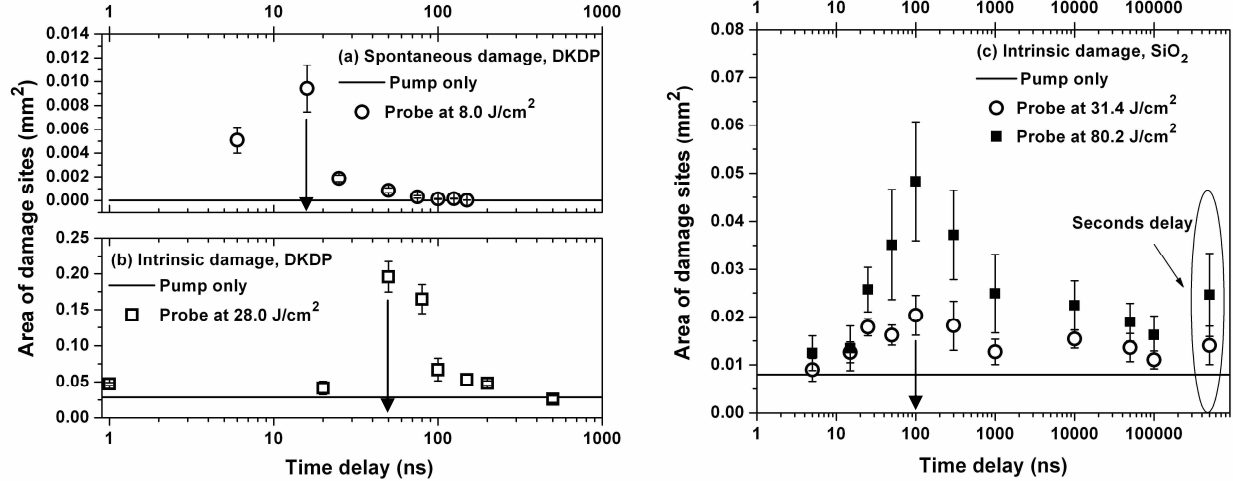


Figure 4. Cross-sectional area of bulk damage sites in the pump and probe interaction region for the case of (a) spontaneous and (b) intrinsic damage in DKDP, and (c) intrinsic damage in SiO<sub>2</sub> as a function of probe time delay for fixed probe fluences. Size of damage from pump only is also shown for comparison (solid lines).

in Fig. 4(c) for two different probe fluences and time delays up to 100  $\mu$ s. In addition, the damage size in SiO<sub>2</sub> corresponding to few seconds (i.e., infinite) probe delay is also shown at  $\sim$ 0.5 ms on the same graph (for illustration purposes), as it is relevant to damage growth. For comparison, the size of damage sites from pump only is plotted as a constant baseline in each graph (solid lines). Throughout this work, individual data points represent the average over measurements performed at five testing locations for each probe delay and fluence combination. The error bars are then derived from the standard deviation of the measurement.

The increase in the final size of damage sites following pump and probe interaction as compared to those from pump only confirms the presence of enhanced absorption by the host material surrounding the location of energy deposition. Figures 4(a)-(c) illustrate several interesting features of the localized dynamics of the transient absorption. Namely:

- i) The size of the damaged region, and thus the amount of energy absorbed from the probe pulse and deposited into the material, first increases with delay time and then decreases. In DKDP, the peak in the absorption profile is located at  $\sim$ 20 ns and  $\sim$ 50 ns, for spontaneous and intrinsic damage, respectively, while the maximum sized damage region in SiO<sub>2</sub> is observed at  $\sim$ 100 ns [indicated by the vertical arrows in Figs. 4(a)-(c)]. Moreover, in the former case, the peak location shifts to longer delays with increasing focal volume (i.e., increasing energy deposition volume).
- ii) The transient absorption of the laser-modified material is longer lived in fused silica, decaying over  $\sim$ 100  $\mu$ s, compared to only  $\sim$ 100 ns observed in DKDP. Even under identical excitation conditions (as is the case for intrinsic damage initiation), the relaxation times in the two materials differ by three orders of magnitude. This behavior, in conjunction with the different peak locations in the absorption profiles of DKDP and silica, as depicted in Figs. 4(b) and 4(c), suggest a strong dependence on the host material properties. This assumption is further supported by the fact that the relaxation time for DKDP is the same for both spontaneous and intrinsic damage [see Figs. 4(a)-(b), under very different excitation conditions].
- iii) For the case of fused silica, the size of damage sites after pump and probe damage testing with seconds delay (i.e., commonly referred to as damage growth with subsequent pulses) appears to be larger than those observed at 100  $\mu$ s time delay. This observation suggests that the mechanisms for additional energy deposition by the probe pulse in the material may be different in the various stages of transient material modifications leading to a damage site (i.e. crack growth may be a contributing factor in the later stages while induced absorption due to localized high temperatures and pressures are playing a role at early times).

The results presented in this study clearly show that the initial energy deposition and resulting plasma modify the surrounding material to become absorbing. This process may occur due to either thermal or mechanical

energy transport from the initial damage event associated with extreme localized conditions of temperatures and pressures and generation of a shockwave. The maximum damaged area in the case of intrinsic SiO<sub>2</sub> damage is about  $5 \times 10^{-2} \text{ mm}^2$  (diameter of  $\sim 250 \text{ }\mu\text{m}$ ) for 100 ns delay. The corresponding velocity is comparable to the speed of sound in silica ( $2.5 \text{ }\mu\text{m}/\text{ns}$  vs  $5.8 \text{ }\mu\text{m}/\text{ns}$ ) while the thermal conduction length is only about  $1 \text{ }\mu\text{m}$  for 100 ns. Thus we assume that the intense pressure pulse following energy deposition (by the pump pulse) modifies the surrounding material as it propagates. As a consequence, the volume of the induced absorbing region increases at early times. At the same time, the shockwave weakens as it spreads and the material modification decays in time. The superposition of the above effects leads to the behaviors depicted in Figs. 4(a)-(c), namely, there is a maximum size damaged region as a function of probe time delay in both materials.

#### 4. SUMMARY

The results of pump and probe damage testing presented in this work have revealed the presence of induced transient absorption within the modified host material well outside the energy deposition volume. We have showed that the response of the material is largely independent of the excitation process and is characteristic of the material properties. The induced absorption can be understood in terms of PdV work done by a shockwave launched from the initial energy deposition by the pump pulse creating defects of different lifetimes in different materials.

#### ACKNOWLEDGMENTS

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