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Optics Performance at 1ω, 2ω, and 3ω: Final Report on LDRD Project 03-ERD-071

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Auspices Statement

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FY05 LDRD Final Report
Optics Performance at 1ω, 2ω, and 3ω
LDRD Project Tracking Code: 03-ERD-071

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Abstract

The interaction of intense laser light with dielectric materials is a fundamental applied science problem that is becoming increasingly important with the rapid development of ever more powerful lasers. To better understand the behavior of optical components in large fusion-class laser systems, we are systematically studying the interaction of high-fluence, high-power laser light with high-quality optical components, with particular interest on polishing/finishing and stress-induced defects and surface contamination. We focus on obtaining comparable measurements at three different wavelengths, 1ω (1053 nm), 2ω (527 nm), and 3ω (351 nm).

Introduction/Background

The interaction of intense laser light with dielectric materials is a fundamental applied science problem that is becoming increasingly important with the rapid development of evermore powerful lasers. For the large fusion-class (i.e. high energy, multi-megajoule, and high power, hundreds of terawatts to petawatts) laser systems, the ability to operate efficiently and reliably at or above their design specifications will ultimately be determined by the performance of their optical components. For these systems, the optical components of interest are the amplifiers (glass or crystal lasing material), frequency converters (potassium dihydrogen phosphate (KDP) and deuterated (DKDP) crystals), transmissive optics (lenses, diffractive optics, and windows), reflective optics (hafnia-silica multilayer mirrors), and polarizing optics that can both transmit and reflect light. Because for a given laser system the irradiation conditions depend on how the laser is operated and thus vary with wavelength, fluence level, and pulselength, it is of particular importance to identify and understand the laser material interactions that lead to damage of the optical components.

From the point of view of the materials, materials are not perfect but rather they contain defects. During the interaction with intense laser light, these defects may provide sites where considerable amounts of energy can be deposited into the material [1-3]. Absorbing type defects within materials cause heating, and potentially, plasma formation. As the hot plasma expands, the surrounding material is both thermally and mechanically stressed and damages. Nonabsorbing defects such as cracks, voids, and lenslets, result in

dielectric discontinuities that also can lead to damage by enhancing the local electric field of an optical wave and/or serve as positive or negative lenses for the incident laser light. This in turn can cause high local power densities to occur leading to damage at lower average fluences than expected when compared to bulk material [4]. Voids and cracks can also serve to trap absorbing impurities [5].

Defects can originate during manufacture of the raw material (trace impurities, inclusions and voids) or be produced during the polishing or finishing process (scratches, surface impurities, subsurface damage such as cracks and residual stress). For mirrors and polarizers, defects can also occur during the deposition process used to create highly reflective multilayer coatings (nodular defects or particles that result from spattering of the starting materials) [6]. Under certain irradiation conditions, some defects lead to damage while others do not. Once initiated, during repetitive illumination damage sites may either be stable (remain small and constant in size) or grow to catastrophic proportions ejecting material to form large craters [7]. To complicate matters, extrinsic factors such a dust or organic vapors that adhere to the surface of optics can further degrade their performance. Consequently, understanding the complex mechanisms that describe the response of optical materials to intense laser irradiation and, in the presence of contaminants, is of crucial importance for operating these high-power lasers at their performance limits.

The effort at LLNL to improve the performance of optical materials under intense illumination in large laser systems has been ongoing for nearly three decades. More recent activities in support of the National Ignition Facility (NIF) laser have been underway since project inception in 1993. Until just recently, the emphasis of this work was directed toward measuring damage parameters at the design basis wavelength of ω, which is more stressing on optical materials. Of particular importance were studies in the following areas:

Lenses, Windows, Diffractive Optics (Fused Silica):

- Laser-induced damage initiated by contaminant particles or organic vapors on surfaces [8-10].
- Initiation of damage as a function of fluence and pulselength for fused silica for a limited number of vendor proprietary surface finishes [11].
- Laser-induced damage initiated at scratches or due to bulk inclusions [12].
- Growth of damage sites as a function of repeated illumination [13].
- Mitigation of small initiated damage sites using CO_2 lasers and mechanical material removal to retard growth with subsequent laser pulses [14].
- Studies of anti-reflective (AR) coatings [15].

Frequency Converters – (doublers - KDP and triplers - DKDP)

- Initiation of damage as a function of fluence and pulselength for doublers [16].
- Bulk damage of tripler crystals as a function of fluence and pulselength [17-18].
- Studies of tripler conditioning for the reduction of bulk damage [19].
- Developing the growth processes for large-scale crystal production. The steady improvements in KDP and DKDP large-scale crystal growth are the result of this effort and have been reported by [20-24].

Most of the fused silica damage initiation and growth studies at 3ω were performed without unconverted 1ω and 2ω light. In large-scale laser systems that employ frequency conversion, multiple wavelengths in various proportions can strike optics simultaneously because the conversion process in not 100% efficient, leaving some unconverted 1ω and 2ω light mixed with the 3ω light. The combined effect of the multiple wavelengths has not yet been systematically studied.

The performance of optical components that only "see" light at 1ωhas also been studied. For example, the functional damage threshold of multilayer polarizers, nodular defects in multilayer mirrors, and the effects of Pt inclusions on possible damage in Nd-doped metaphosphate amplifier slabs (laser glass) have been investigated [25-26].

Research Activities

The research effort focused on two activities: 1) systematic studies of damage initiation and growth for fused silica as a function of contaminant, fluence and wavelength; 2) studies related to KDP and DKP, specifically a) enhancing understanding of the damage initiation mechanisms and their consequences, b) examining pinpoint density and size as a function of wavelength, fluence and pulse length, c) improving the performance of KDP and DKDP crystals through conditioning.

Results/Technical Outcome

Laser damage growth in fused silica

Laser damage growth in fused silica with 1ω and 3ω

Optics after the frequency converter crystals will see 3ω and unconverted red and green light. The residual green is negligible, but the unconverted red light can be appreciable depending on the operating conditions. Measurements taken to look 3ω operation, at fluences less than the growth threshold for 3ω only, are summarized in Figure 1. Here the exponential growth coefficient, alpha, is plotted vs. the sum of the 1ω and 3ω fluence. The diameter, d, of a damage site grows exponentially with the product of alpha and N, the number of shots, or $d = d_o * \exp(\text{alpha}(F) * N)$. Also shown on this plot is the least squares fit to the previously measured 3ω growth. Two fluences at 3ω are plotted here; one approximately 30% lower than the 3ω growth threshold and the other just at threshold. Until the residual red reaches approximately 5 J/cm^2 growth is not triggered. Baseline converter calculations predict that for 3 ns pulses the unconverted light should be in the 2 J/cm^2 range and thus should provide no additional threat. The same calculations predict that for 10 ns or longer pulses, the unconverted light will be in the 6 J/cm^2 and thus will pose a threat.

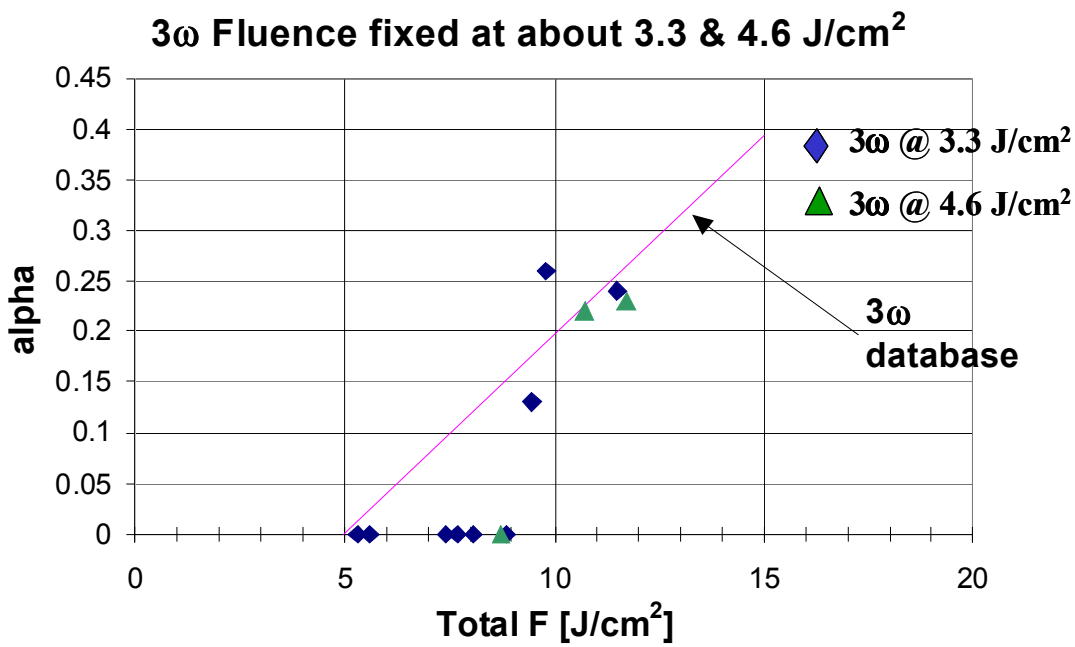

Figure 1. Growth of laser damage in fused silica with combined 1ω and 3ω.

Laser damage growth in fused silica with 1ω and 2ω

With the FOA optimized for frequency conversion into 2ω, there will be unconverted red light. The residual red expected depends on the installed converter crystals and unconverted red light can be appreciable for the thin doubler used with the tripler. Measurements taken to date to look at 2ω operation at fluence less than the growth threshold for 2ω only are summarized in figure 2. Here the exponential growth coefficient is plotted vs. the sum of the 1ω and 2ω fluences. Also shown on this plot is the least squares fit to the previously measured 2ω growth and 3ω growth data. Three

fluences at 2ω are plotted here; all are less than the observed 2ω growth threshold of 12 J/cm^2. The no growth data points are slightly offset from 0 to facilitate distinguish the 2ω fluence. Again, we rely on predicted calculations for the NIF converter to assess the threat of the residual red. For the thin, 13 mm thick doubler crystal, the residual red at 3.5 ns and longer pulse widths may trigger growth for operation below threshold for green alone. For the proposed 18 mm thick doubler, the residual red for operation at 9 J/cm^2 of green would be expected to have little impact.

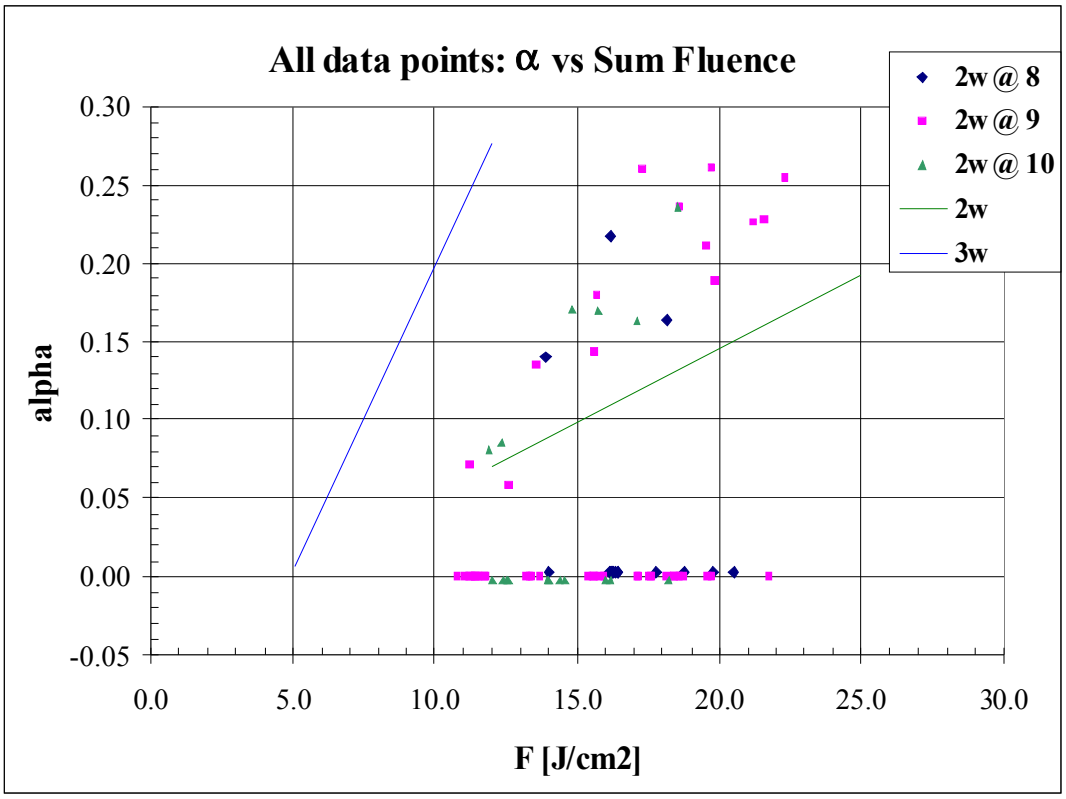

Figure 2. Growth of laser damage in fused silica with combined 1ω and 2ω.

These data along with 1ω +3ω growth rates were presented recently [27].

Laser damage growth in fused silica with 1ω only

We have extended our single wavelength database for growth of laser-initiated damage in fused silica to 1ω. The data taken at all 3 wavelengths is shown in Figure 3. Here we note that when growth is triggered to start at 1ω fluences above 20 J/cm^2 it grows faster than that measured at 2ω and, furthermore, the growth rate depends less on the fluence than the dependence observed at the two shorter wavelengths. Work on 1ω growth is continuing.

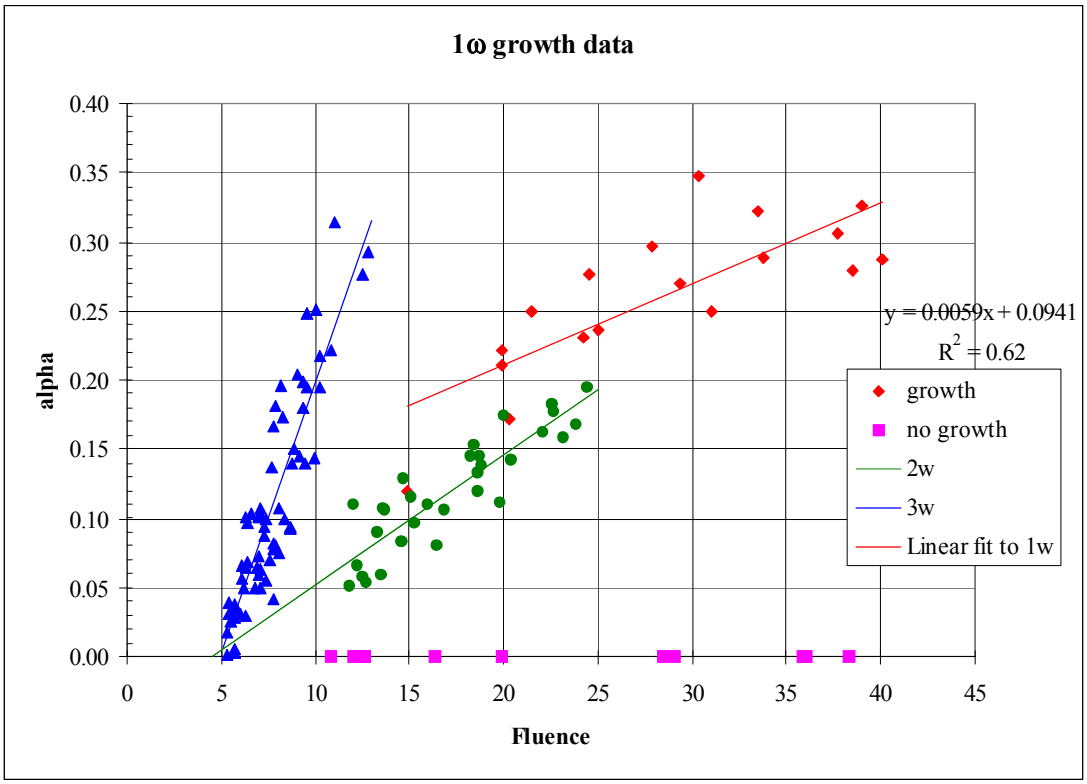

Figure 3. Growth of laser initiated damage in fused silica at all 3 wavelengths.

Studies related to KDP and DKDP

Enhancing understanding of damage initiation mechanisms

Forensic studies of bulk damage sites, which appear in KDP and DKDP crystals, have led to enhanced understanding of both the mechanisms of damage initiation and it's consequences for operation in large, high energy, high-power laser systems. Possibly the most fundamental advance of these studies comes from our improved understanding of the morphology of an individual damage site. It has been well known for many years that laser-induced damage in KDP and DKDP crystals manifests as small, localized scattering centers distributed thought a beam footprint. These scattering centers are often referred to as pinpoint damage sites or simply pinpoints. Recently scanning electron microscopy (SEM) has been used to image individual pinpoints at unprecedented levels of detail. Figure 4 shows a dark field image of bulk damage in a beam footprint and an SEM image of an individual pinpoint.

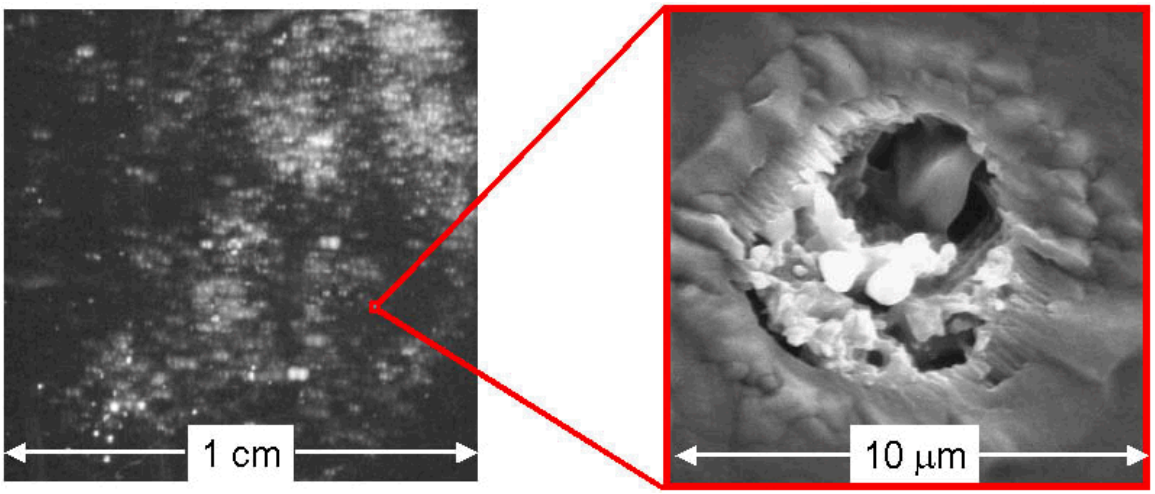

Figure 4. Left: A field of bulk damage produced by an 8-J/cm^2 3-ns 3ω laser pulse in unconditioned material. Right: A SEM image of an individual bulk damage site. The image was obtained by breaking a piece of DKDP though the damage site so that it could be examined with SEM and other techniques typically restricted to surface analysis.

Historically it has been assumed that in the presence of multiple wavelengths that the light of the shortest wavelength would dominate damage initiation (creation of pinpoints) because of the consistently lower damage thresholds observed at shorter wavelengths.[28,29] During the high-3ω-fluence campaign on NEL (NIF Early Light), a hot spot was introduced into the beam by a defect in the PABTS (Pre Amplifier Beam Transport & Steering). This high fluence region produced a matching region of enhanced damage density in the third harmonic generator. Closer examination of the damage density as a function of depth, ρ(z), produced an unexpected result; most damage was heaviest in the center of the optic.[30]

To understand why this result was surprising we must first consider how the fluence of each wavelength varies a function of depth in the THG Third Harmonic Generator, a DKDP crystal). Light incident on a THG has is composed of 1/3 1ω light and 2/3 2ω (as measured by fluence). As the light propagates through the crystal 1ω and 2ω photons are combined to produce 3ω light with some residual of the other two colors. As this process occurs over the entire depth of the crystal the fluence of each wavelength will vary with depth in the crystal. Historically the damage density would have been expected to be heaviest at the depth where the 3ω fluence was highest. Figure 5 shows a) the calculated fluence of each wavelength as a function of depth and b) the observed damage density as a function of depth.[31] The maximum 3ω fluence is calculated to be at z = 8 mm (as measured from the input surface), yet the peak of the damage density is closer to z = 4.5 mm.

While initially puzzling, we found that the damage density could be readily described by assuming that damage was initiated through an interaction of 2ω and 3ω light, but not 1ω light. This was significant for two reasons. Historically, damage test data has been taken

during single wavelength experiments. These results indicated that the presence of 2ω light could not be discounted, prompting various multi-wavelength damage experiments to be performed. On a more fundamental level, the interaction of the 2ω and 3ω light (but not the 1ω) suggested that the defect-assisted multi-photon damage initiation model for damage initiation proposed by Carr and coworkers was also applicable in the presence of multiple wavelengths. [28]

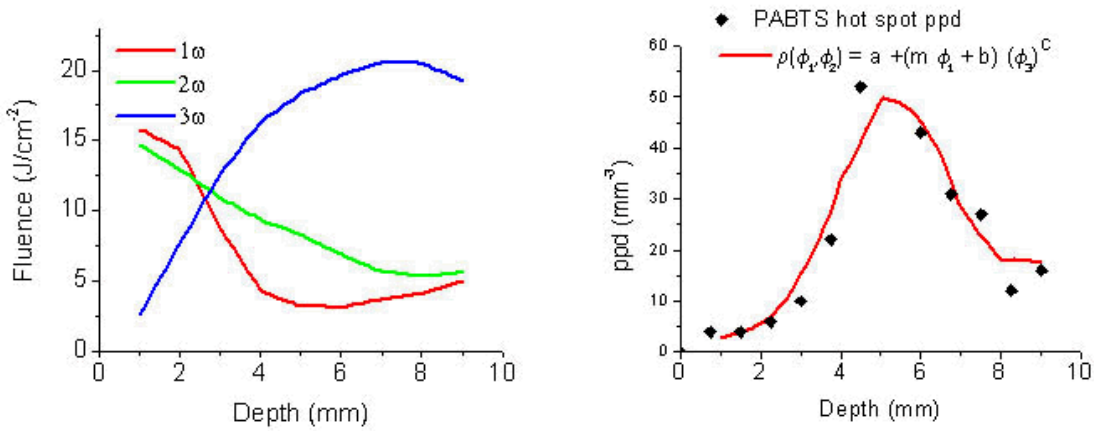

Figure 5. Left: The fluence as a function of depth in the PABTS hot spot for 1ω, 2ω, and 3ω light. Right: solid points indicate the pinpoint density as a function of depth at the PABTS hot spot. The solid red line indicates the fit to pinpoint density assuming the 2ω and 3ω (but not the 1ω) fluences contribute to a multi-photon damage initiation mechanism.

In addition to an enhanced understanding of the mechanisms by which damage is initiated in converting crystals we have made improvements to understanding the consequences of bulk damage in SHGs (Second Harmonic Generator, typically a KDP crystal) and THGs. As bulk damage sites in KDP and DKDP are not observed to grow at fluences below 20 J/cm^2, obscuration by pinpoints and its addition to beam contrast is the metric by which the bulk performance of crystalline parts is gauged. To minimize the effect of bulk damage on beam contrast, a set of frequency converters may not have a contrast adder larger than ~5%. A 5% contrast adder corresponds to 0.1% of the beam energy being scattered. The maximum allowable number of pinpoints (the number which will scatter 0.1% of the incident light) is determined by their size and opacity. For unconditioned crystals pinpoints produced by 3-ns 3ω light are believed be on order of 5 μm in diameter.

Figure 6 shows a bright field image of a pinpoint and the luminosity counts through both its core and through an area of equal size not including any damage. The integrated luminosity in the core of the pinpoint is half that of the undamaged region. And thus pinpoints are considered to be 50 percent opaque for the purposes of energy scatter.

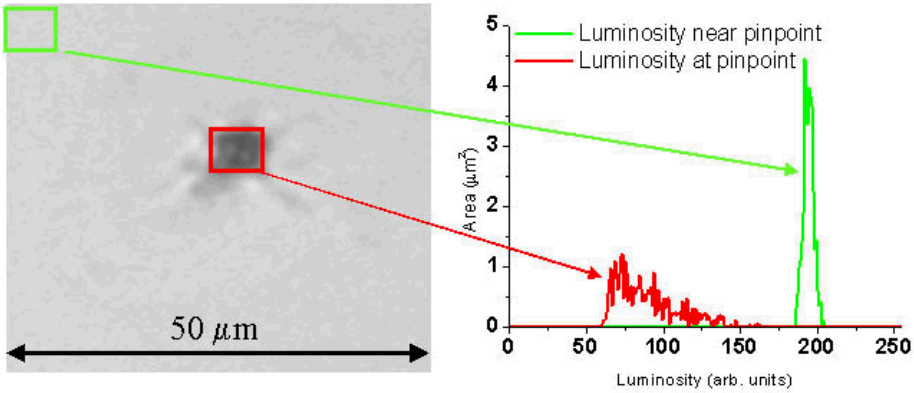

Figure 6. A bright field image of a pinpoint and the luminosity counts through both its core and through an area of equal size not including any damage.

There is some question whether the region outside the core will contribute significantly to the scatter caused by the pinpoint. SEM images such as in Figure 4 suggest that the haze just out side the core of the pinpoint is material modified during the damage process. First cleaving the sample open and then shocking it can separate the shell of modified material from the surrounding bulk (see Figure 7).

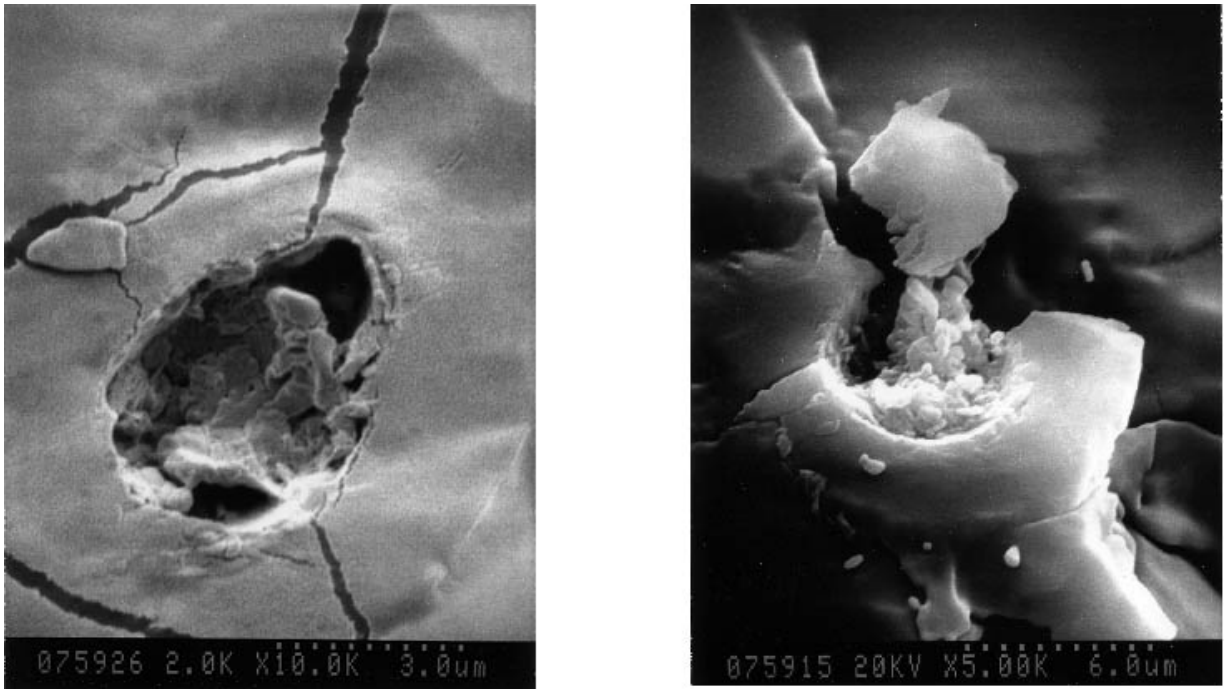

Figure 7. The shell of modified material immediately adjacent to the pinpoint is seen to separate from the surrounding bulk material after a shock. The lack of wake hackle indicates that the fractures observed formed after the crystal was cleaved. The dark annulus surrounding the crater on the left indicates a region of

reduced electrical conductivity. The reduced electrical conductivity may be due to the modified material or due to a reduced adhesion of the palladium coating (necessary to obtain SEM photos) to the modified material.

Recent measurements in DKDP may suggest that the shell of material surrounding the pinpoints core is also chemically different than the bulk marital. Figure 8 shows a microscopic image of a bulk damage site in DKDP and its associated Raman spectra. The reduction of some modes and the enhancement of others may indicate either structural or chemical change in the shell of material surrounding the core of the pinpoint.

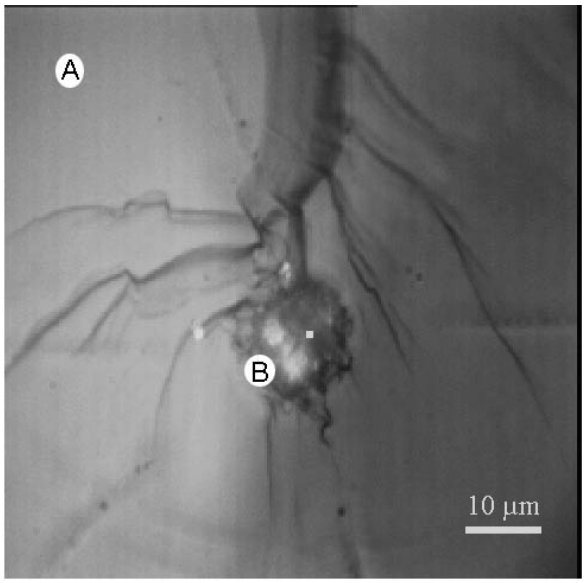

Figure 8a). An optical microscope image of a bulk pinpoint in DKDP produced by 1ω light. The larger core size in this image (as compared to Fig. 4) is typical for the difference between damage induced with 1ω and 3ω light.

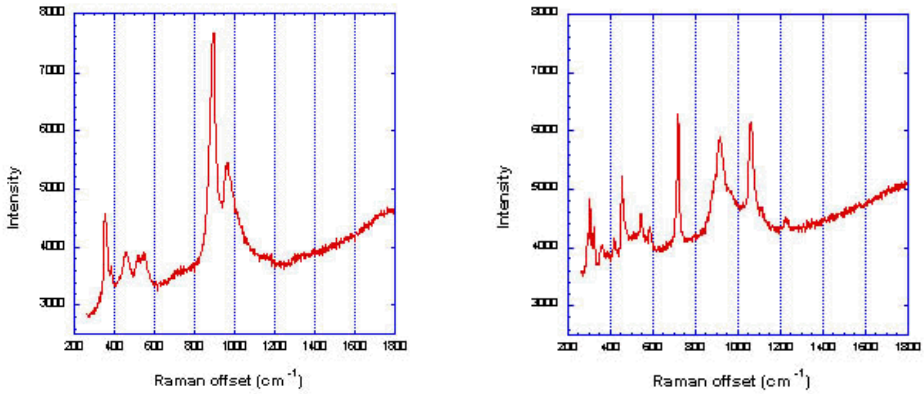

Figure 8b). Left: Raman spectra obtained at site A. Right: Raman spectra obtained at site B. The enhanced modes in the right had spectra could indicate physical or chemical change in the core surrounding the pinpoint.

Examining the enhancement and reduction of these various modes with two-dimensional micro-Raman mapping reveals a structure similar to that observed in the SEM images in Figures 4 and 7. Figure 9 indicates that the 900 cm^{-1} mode (associated with the PO_4 stretch) is depressed in the pinpoint core. The enhancement of the 716 cm^{-1} mode correlates well to the shell around the core observed in the SEM images in Fig. 7.

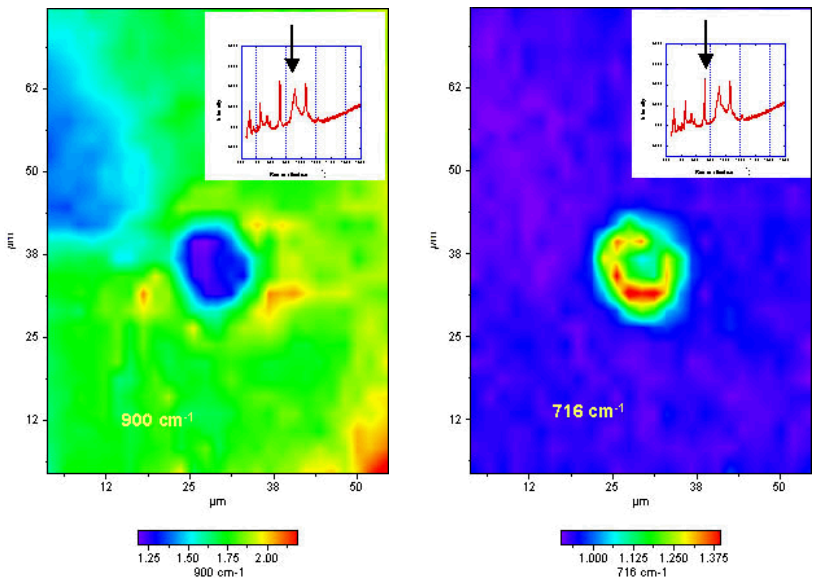

Figure 9. (plots by M. Feit) Left: Indicates a suppression of the 900 cm^{-1} peak in the vicinity of the crater. Right: A new/enhanced peak at 712 cm^{-1} is visible around the rim of the crater, potentially corresponding to the shells observed in Fig. 7.

As damage sites have been measured to reach temperatures on the order of 10,000 K and correspondingly high pressures, it is not surprising to find melted and or compacted material surrounding the damage site. [32]

Pinpoint density measurements as a function of pulse length

Measurements were made using the Nexiv microscope on various sites in LL11-1A that were unconditioned and had been single-shot damage tested on OSL at 0.1, 0.3, 0.8, 1, 3, 10, and 20 ns. The Nexiv microscope was used to automatically count and size each bulk damage site through the volume of the crystal that had been damaged. The size reported is the calculated semi-major axes of an ellipse that best fits the bulk pinpoint shape. Figure 10 shows a graph of the calculated average size of the bulk damage sites at each of the volumes that had been damaged. Both dimensions of the pinpoints are shown. We do not see a large difference between the perpendicular sizes of the pinpoints from the graph. However, inspection of the numerical data suggests that the pinpoints are elliptical with an eccentricity of 0.75.

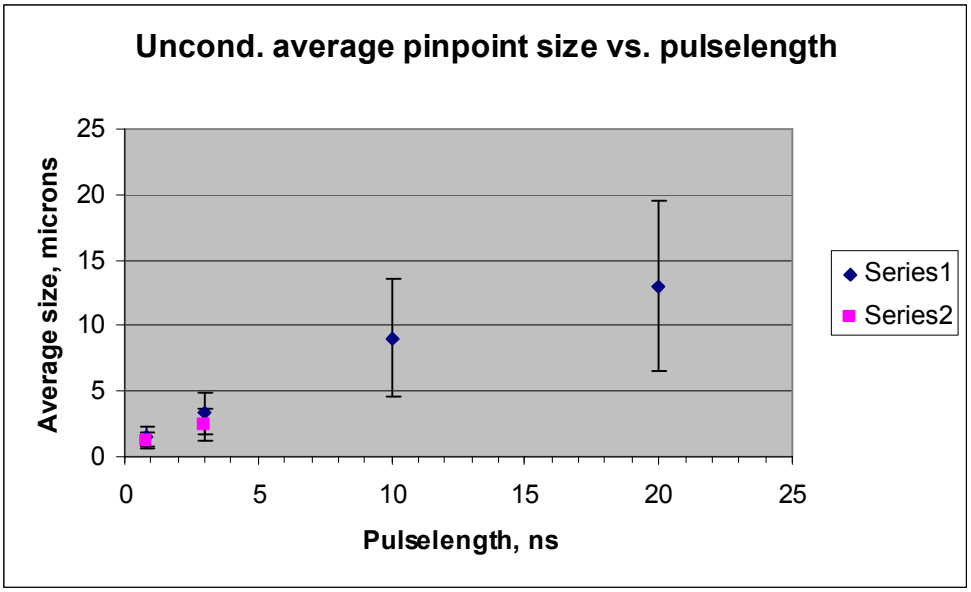

Figure 10. Average dimension of a bulk damage pinpoint as a function of damaging laser pulse width. Series 1 and Series 2 are the values for the semi-major axes from the pinpoints fitted ellipse.

Improving performance of DKDP crystals through conditioning

The performance of an optic at a given fluence, ϕ, can often be described by ρ, the number of damage sites per unit area. As pinpoint damage is usually the greater concern in DKDP, ρ in this case is defined as the number of damage sites per volume. Some $\rho(\phi)$ curves were measured for LL16 and LL14 tripler cut DKDP for several conditioning pulse widths tested at 3ω, 3ns. The performance of the $\rho(\phi)$ curves for the conditioned material was compared to $\rho(\phi)$ for unconditioned material in terms of a figure of merit. Our figure of merit is FOM = (ϕ_{10} for conditioned material/ϕ_{10} for unconditioned material) x 100. When we plot FOM vs. conditioning *intensity* we find an interesting relationship. Figure 11 shows that the relation between conditioning FOM and conditioning intensity in our range of intensities is linear. Also, we see that 300-ps conditioning at 10 GW/cm^2 provided the best conditioning effectiveness. Conditioning results for KDP are presented in [33].

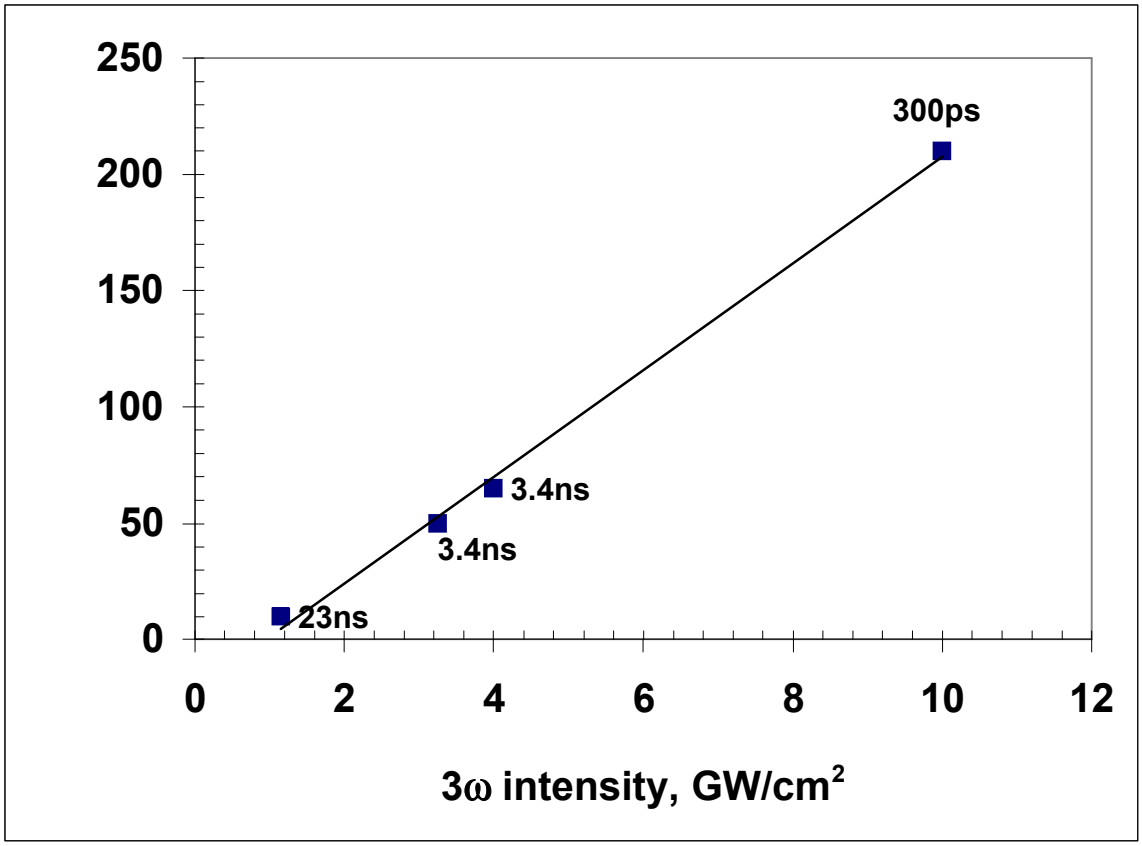

Figure 11. Conditioning effectiveness (FOM) vs. conditioning intensity

Exit Plan

At the conclusion of this ER, we have significantly enhanced our fundamental understanding of laser material interactions in terms of wavelength, fluence, pulselength, and size, density, and type of defects that result in the performance degradation of optical components in fusion-class lasers, particularly with regard to fused silica and KDP/DKDP crystals. Through this increased understanding, we have already begun to develop novel conditioning methods that will retard the onset of damage and minimize its growth once initiated. Current models of damage initiation and pulse length scaling will be extended to connect hypothetical distributions of absorbing defects directly with measurable quantities such as damage initiation probabilities and beam obscuration for surface and bulk damage in both fused silica and KDP/DKDP. As part of this project, we have also developed a working, living stochastic model of the evolving damage state of a fusion class laser system that can be used to predict the useful life of optical components.

Acknowledgement
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