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Optical scatter as a diagnostic tool for studying bulk defects which cause laser damage in conventional and rapid growth KDP and DKDP

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I. INTRODUCTION

Single crystals of KH_2PO_4 (KDP) and $(D_xH_{1-x})_2PO_4$ (DKDP) will be used for frequency conversion and as part of a large aperture optical switch in the proposed National Ignition Facility (NIF) at the Lawrence Livermore National Laboratory (LLNL). These crystals must have good optical properties and high laser damage thresholds. Currently these crystals have a lower laser damage threshold than other optical materials in the laser chain which has forced designers to limit the output fluence of the NIF in order to avoid damaging the crystals. Furthermore, while more efficient frequency conversion schemes are being explored both theoretically and experimentally, the advantages of these schemes can not be fully realized unless the damage thresholds of the conversion crystals are increased. Over the past decade, LLNL has generated an extensive data base on the laser damage in KDP and DKDP crystals both at the first and third harmonics of Nd-YAG.[1] While the damage thresholds of these crystals have increased over this time period due, in part, to better filtration of the growth solution,[2] the damage thresholds of the best crystals are still far below what is expected from theoretical limits calculated from the band structure of perfect crystals. Thus damage in KDP and DKDP is caused by defects in the crystals. We also rely on a process called laser conditioning to improve the damage thresholds of the crystals. Unfortunately, little is understood about the mechanism of laser induced damage, the conditioning process in the crystals, or the defects which are responsible for damage. We have recently implemented a scatter diagnostic for locating and studying defects in crystals and as a tool for studying the mechanism of laser damage and laser conditioning.

II. LASER DAMAGE

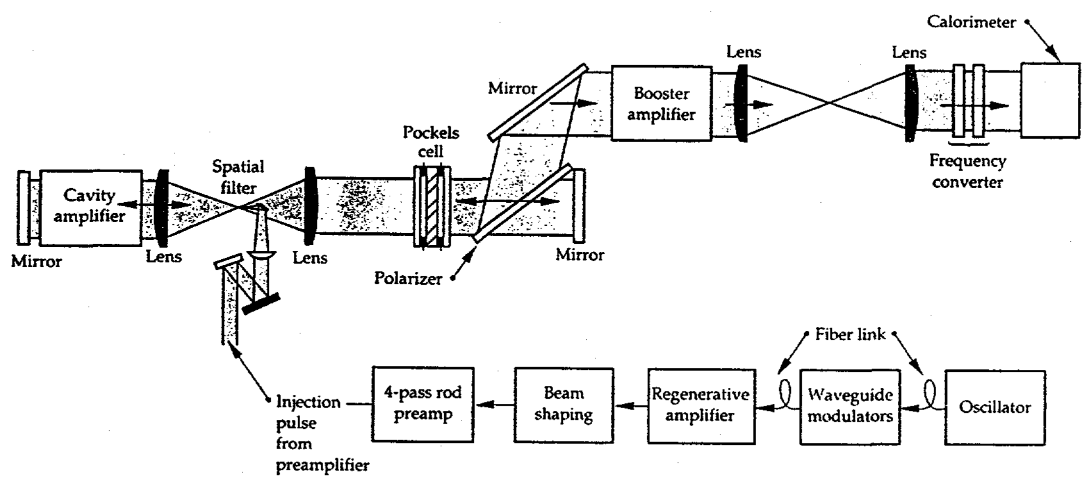

Figure 1 Beamlet Laser System

Figure 1 shows a schematic layout of the Beamlet laser system.[3] Beamlet is a scientific prototype of a single beamline for the NIF. KDP is used in the Pockels cell which acts in conjunction with a polarizer to form a large aperture optical switch which can contain a pulse of light in the main laser cavity for multiple passes through the amplifiers or switch the light out of the main cavity after it has taken multiple passes through the amplifiers.[4] In Beamlet the light is switched out of the main cavity after four passes through

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the amplifiers. KDP is also used for the frequency doubler to convert to the second harmonic. DKDP is used for the frequency tripler to convert to the third harmonic. We use DKDP in this location because the gain for stimulated Raman scatter (SRS) is substantially lower than for KDP at the third harmonic.[5]

The NIF requirements for the laser damage thresholds at each location where the crystals will be used and the current laser damage thresholds for KDP and DKDP for crystals being used on Beamlet are shown in Table 1. S-on-1 refers to test conditions where each site is subjected to 600 shots at the same fluence. The sample is then viewed under an optical microscope where any visible change 10μm or larger would constitute optical damage. For R-on-1 tests each site is subjected to a ramped fluence starting at a value well below the nominal damage threshold and steadily increasing to the final test fluence for that site. As can be seen from the data, R-on-1 laser damage thresholds are higher than S-on-1 values. This is a well known phenomenon — but not a well understood one — called laser conditioning.[6] The laser damage thresholds for the Pockels cell crystal and the frequency doubler are well within the requirements for the NIF for both S-on-1 and R-on-1 at 1064nm (1ω). The frequency tripler, however, does not currently meet the NIF requirements at 355nm (3ω) unless the crystals undergo a conditioning process. Furthermore, the component that operates nearest its laser damage threshold in the current NIF laser design is the frequency tripler. Also the variability in the quality of crystals is such that although most are quite good we are not guaranteed that all will meet the laser damage requirements for the NIF. For these reasons we have initiated a program to study the mechanisms of laser induced damage and the conditioning process.

Location / Material / Wavelength	Damage threshold S-on-1	Damage threshold R-on-1	NIF requirements
Optical Switch KDP 1ω	30	40	14.9
Frequency Doubler KDP 1ω	30	40	15.0
Frequency Tripler DKDP 3ω	10	20	12.7

Table 1 Damage threshold and NIF requirements for KDP and DKDP in J/cm^2 at 3ns

Historically the laser damage thresholds have improved as a result of better solution preparation and growth processes. Figure 2 is a plot of laser damage thresholds as a function of year grown at 1ω and 3ω for both conditioned (R-on-1) and unconditioned (S-on-1) test conditions. It is evident that the damage thresholds have increased over time. It should be noted that this plot includes all damage tests on all crystals so the median values one can deduce from the plot are not representative of the best damage thresholds that are currently achieved and for crystals used on Beamlet. For example the nominal damage threshold values for KDP crystals that vendors have produced for the Beamlet laser system are given in Table 1 as 30 and 40J/cm^2 for S-on-1 and R-on-1 respectively. While these thresholds could be controlled by typical defects in high quality crystals such as point defects (impurity atoms) or dislocations, the improvement in damage thresholds in response to better filtration[2] suggests that inclusions play a significant role in damage.

III. SCATTER MEASUREMENT

Figure 3a is a schematic of the scatter measurement that we have implemented to view defects and bulk damage in the crystals. We have installed the scatter diagnostic in the ZEUS laser damage facility at LLNL. The ZEUS system is capable of delivering a usable high fluence beam of 100J/cm^2 at 1ω and 75J/cm^2 at 3ω in an 8ns pulse. The crystal is mounted in an X-Y stage that allows the damage beam to be positioned at any point on the crystal. The translation stage is also attached to a rotary stage that allows the test region of the sample to be viewed under an optical microscope using Nomarski or backlighting techniques to detect damage. This is the typical method used to damage test samples. A probe laser is aligned collinear to the damage beam with the aid of an alignment camera. An image of the scattered light from the probe laser is obtained by looking horizontally through the edge of the crystal using a high dynamic range CCD camera (1024x1024 pixel, 14 bit) with appropriate imaging optics. Current system magnification is approximately 2X resulting in each pixel mapping to approximately a 10μm^2 area in the crystal.

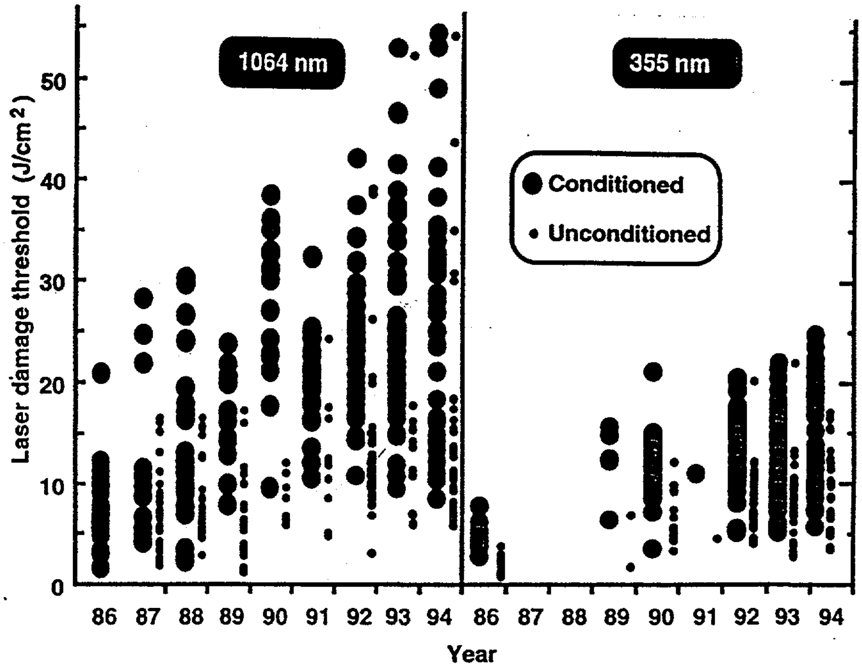


Figure 2 Measured bulk damage thresholds for KDP and DKDP scaled to 3ns values by $\tau^{0.5}$

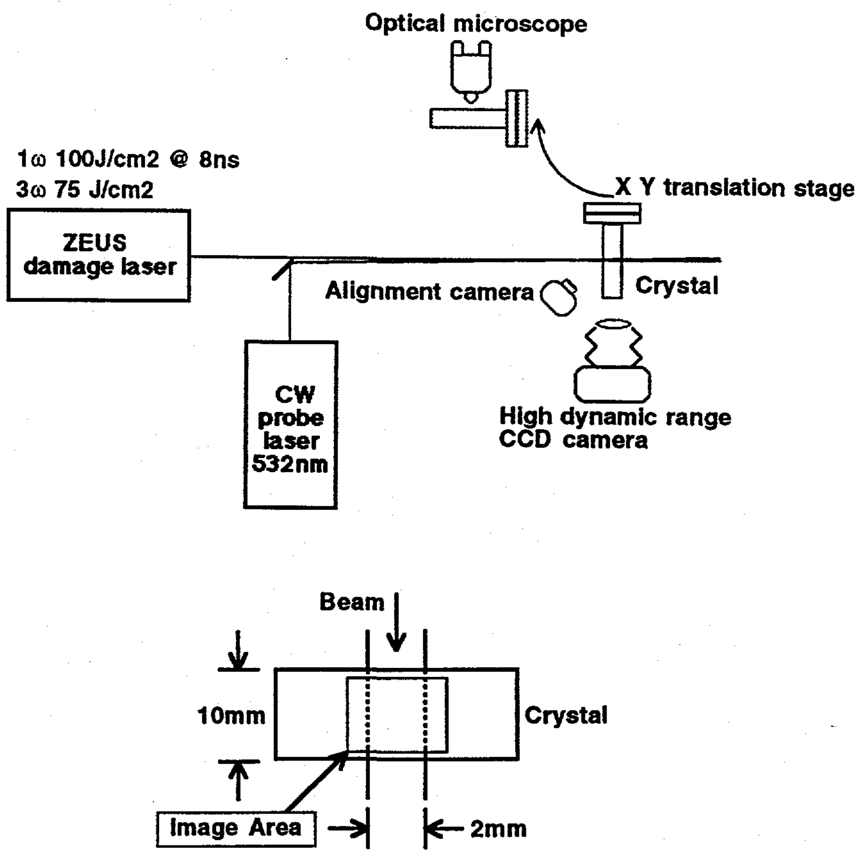


Figure 3 a) scatter measurement system b) schematic of image orientation

Images that are presented in this paper show a scatter track about 8mm long with the beam traveling from top to bottom on the image as shown in Figure 3b. The nominal beam diameter for the ZEUS damage laser is 1mm at $1/e^2$ and the diameter of the probe beam is 2mm at $1/e^2$. We estimate that we can coalign the two beams to approximately ±250μm at the sample plane. The scatter from the surface has to be avoided because it is much brighter than the bulk scatter and will saturate the camera. Signals that we observe consist of several components, those being Rayleigh scatter, Brillouin scatter, Mie scatter, and fluorescence or some nonlinear process that radiates at a different wavelength that the probe beam. The Becke line test has shown that all of the large (Mie) scattering sites that we have observed are regions with an index of refraction that is less than the bulk crystal. We have been able to detect many more defects with this scatter diagnostic than with conventional optical microscopy and have seen dramatic differences between different vintages and types of crystals.

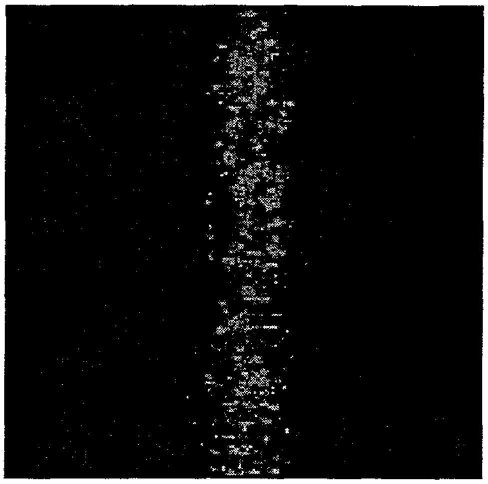
IV. RESULTS

Four different types of crystals having four different growth histories are currently being studied. The first types of crystals were grown before continuous filtration of the growth solution was shown to increase damage thresholds.[2] These crystals are currently being used on the Nova laser at LLNL. Figure 4a is a typical scatter signal for this type of crystal. Many large (Mie) scatterers are visible in this type of crystal. Note that the minimum and maximum values for the gray scale are not the same from image to image in order to highlight important features in each of the images.

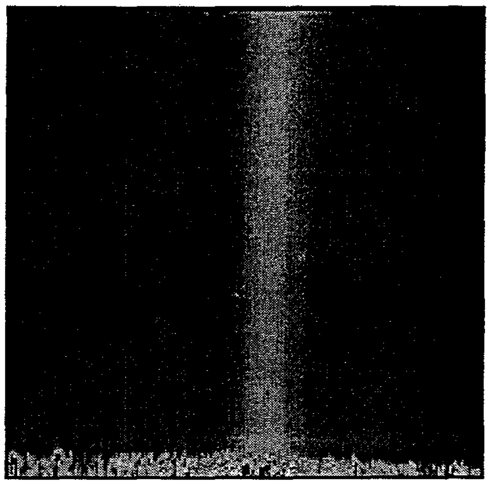
The second types are also being used on the Nova laser at LLNL, but were grown later using better solution preparation techniques. Note that these crystals, see Figure 4b, have substantially fewer large (Mie) scattering sites than earlier. They also have a higher laser damage threshold.

The third types are the most recent crystals grown. These crystals are currently being used on the Beamlet laser system both in the Pockels cell and as frequency converters. These crystals have almost no large (Mie) scattering sites, see Figure 4c, and have the highest laser damage thresholds of any crystals tested to date.

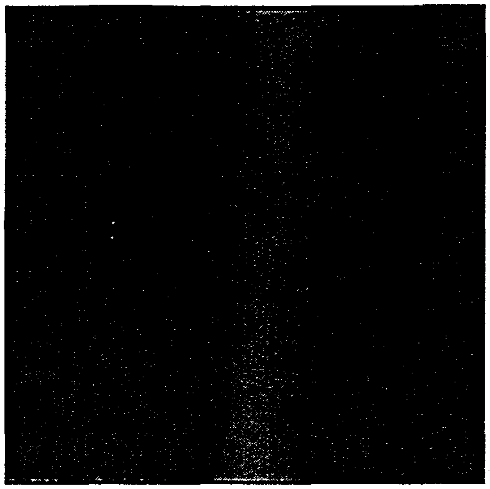
The final types of crystals being studied are fast grown crystals that have been grown at LLNL by researchers who are developing an innovative process for growing KDP and DKDP crystals with excellent optical properties at growth rates up to 10x faster than conventional techniques.[7] This process includes filtration of the solution prior to the start of crystal growth. As is evident from the scatter signal in Figure 4d, these crystals have numerous larger scattering sites which are typically larger in size than the early Nova crystals but are fewer in number.



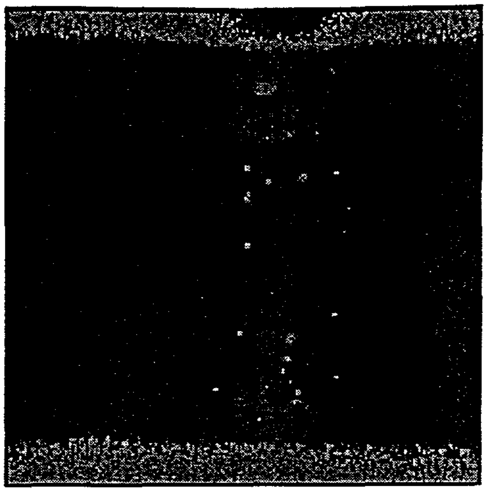
a) Early Nova



b) Late Nova



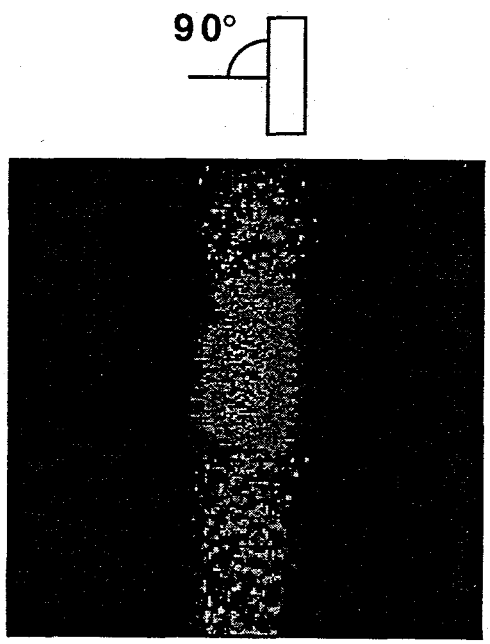
c) Beamlet



d) Fast grown (prefiltration only)

Figure 4 Scatter signals from a) Nova pre-continuous filtration b) Nova post-continuous filtration
c) Beamlet post-continuous filtration d) Rapid grown

We have found scatter to be quite sensitive to the angle at which you illuminate the sample and the angle at which you view the scatter signal. Figure 5 shows a scatter image where the crystal is illuminated at both 90° and 95°. The scatter signal from the region near the center of the image all but goes away when the crystal is tipped by 5°.



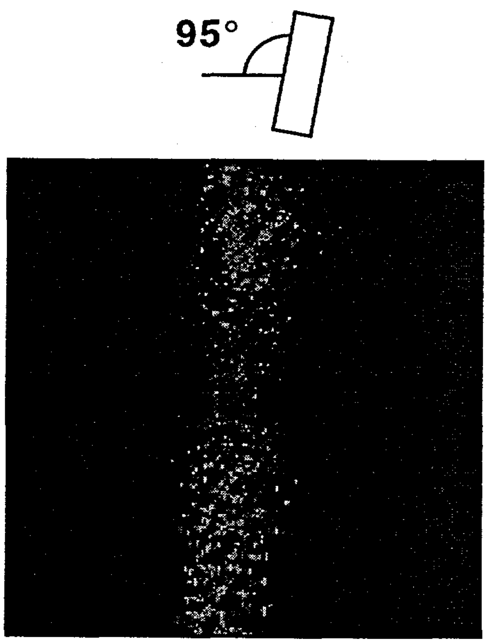


Figure 5 Scatter signal from crystal at normal incidence and at 5° from normal

We have recently seen that the scatter signal is also quite sensitive to the polarization direction of the probe beam. Figure 6 shows a scatter signal from the same location in a Nova pre-continuous filtration crystal for a horizontally polarized probe beam and a vertically polarized probe beam. The vertically polarized probe beam shows many more scattering sites than the horizontally polarized beam. In addition the strength of the Rayleigh signal is greatly enhanced for a vertically polarized probe beam. It is not always the case that a vertically polarized beam results in the observation of more scattering sites. Sometimes horizontal polarization results in more visible scatter sites or in better contrast due to a reduced Rayleigh scatter signal.

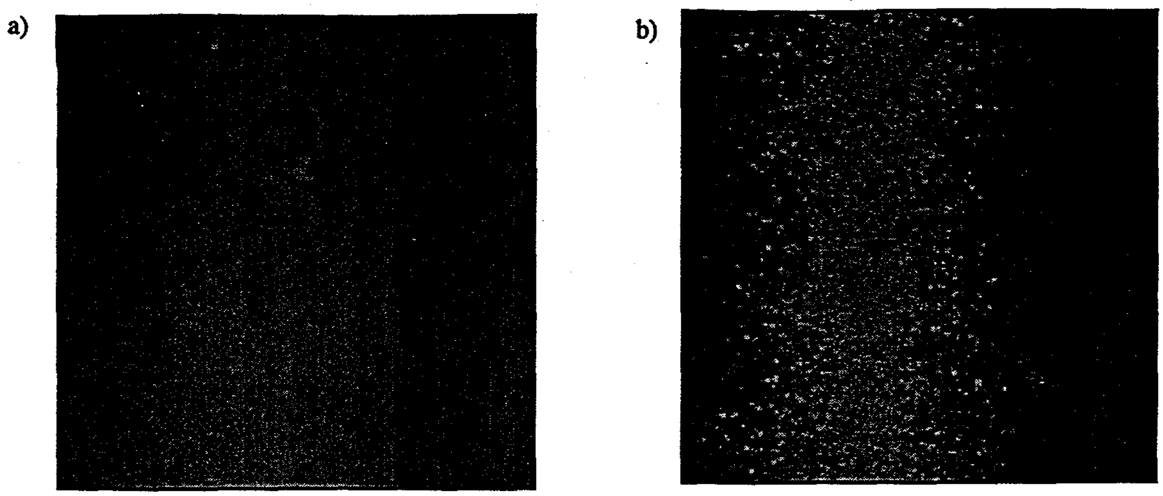


Figure 6 Scatter signal for a) horizontally polarized probe beam and b) vertically polarized probe beam

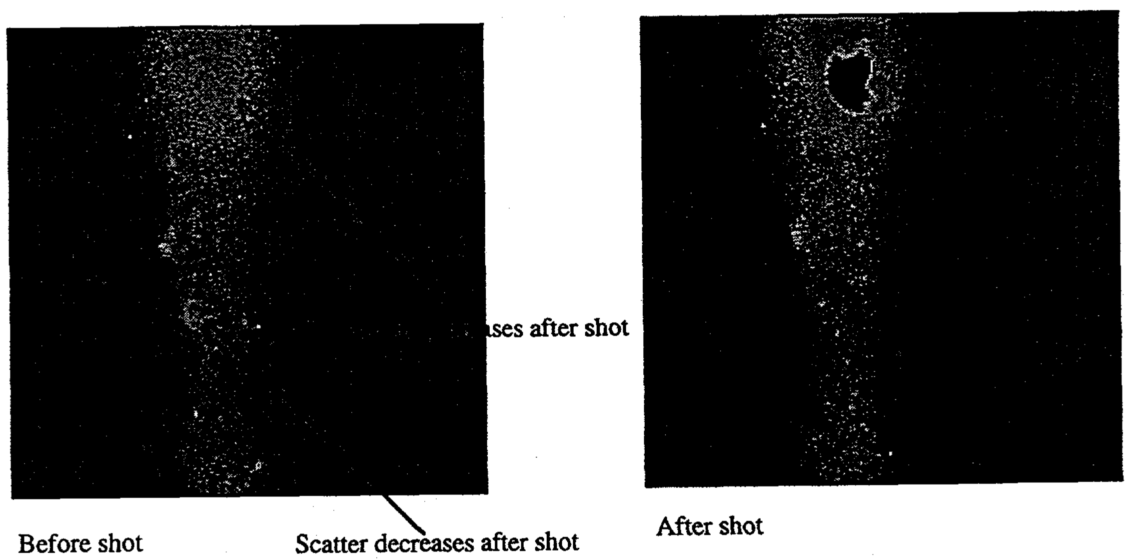
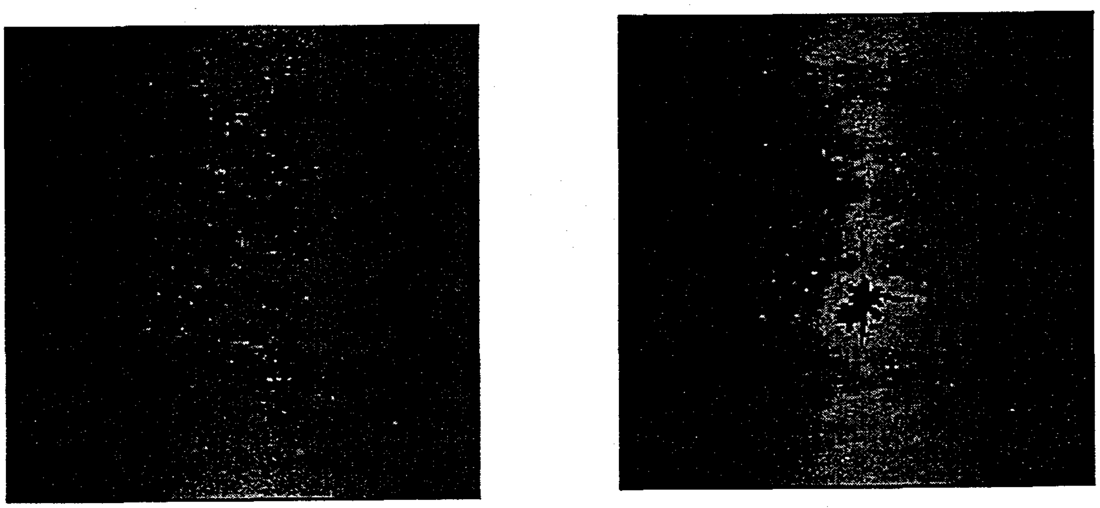


Figure 7 Scatter signal before and after being subjected to subdamage threshold shots

During laser damage tests we have seen several phenomena using the scatter diagnostic system. The signal from some scatter sites increases or decreases dramatically when subjected to subdamage threshold fluences. Figure 7 shows one site whose scatter signal decreased dramatically when subjected to shots at 10.2 and 11J/cm^2. We have seen this decrease in scatter signal when subjected to subdamage threshold fluences occur numerous times and this may be our first glimpse at the conditioning process. We have also seen some scattering sites which initiate laser damage, also shown in Figure 7. However, what we have typically seen is that when laser damage occurs it does not initiate at scatter sites that we had previously observed. This is illustrated in Figure 8. This was an unexpected result and raises the question as to why damage seems to initiate in regions where we don't see initial scatter sites. There are several possibilities. First, the damage may initiate from features which are below our spatial or intensity resolution. We intend to increase the magnification of our detection system to address this possibility. This may also help develop insight into the conditioning process and determine why the amplitude of some scatter sites decreases dramatically when subjected to a high fluence beam. The second possibility is that the correct illumination angle, viewing angle, or polarization direction of the probe beam has not been optimized to detect the sites where damage initiates. We will be conducting a series of experiments to look at different illumination and viewing angles and will begin using circularly polarized light for the probe beam. A third possibility is that damage may initiate downstream from scattering sites due to self focusing. We are examining theoretically whether this explanation is possible. Finally, damage could initiate from regions that have high stress fields due to defects. We plan to develop micron size strain maps for the crystals and correlate them to optical damage.



Before shot

After shot

Figure 8 Images showing heavy laser damage initiating in areas that had no initial scatter sites

V. CONCLUSION

On the ZEUS laser damage facility at LLNL we have installed a scatter diagnostic which allows us to view scatter and optical damage in bulk KDP. We have seen a correlation between level of filtration, number of scattering sites, and optical damage. The amplitude of the scatter signal from large (Mie) scatter sites can increase or decrease when subjected to a laser pulse. Sites whose scatter signal decreases may be providing our first glimpse into the conditioning process. Damage seems to most often initiate in regions where no initial scatter site is observed. We are currently examining this phenomenon with more detailed experiments.

VI. ACKNOWLEDGMENTS

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