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and Pulsed-Laser Irradiation

J. Swain
S. Stokowski
D. Milam
F. Rainer

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Improving the Bulk Laser-Damage Resistance of KDP

by Baking and Pulsed Laser Irradiation*

J. E. Swain, S. E. Stokowski, D. Milam and F. Rainer

Lawrence Livermore National Laboratory

P.O. Box 5508

Livermore, California 94550

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Isolated bulk damage centers are produced when KDP crystals are irradiated by 1-ns 1064-nm pulses. We have tested about 100 samples and find the median threshold to be 7 J/cm^2 when the samples are irradiated only once at each test volume (1-on-1 tests). The median threshold increased to 11 J/cm^2 when the test volumes were first subjected to subthreshold laser irradiation (n-on-1 tests). We baked several crystals at temperatures from 110 to 165°C and remeasured their thresholds. Baking increased thresholds in some crystals, but did not change thresholds of others. The median threshold of baked crystals ranged from 8 - 10 J/cm^2 depending on the baking temperature. In crystals that had been baked, subthreshold irradiation produced a large change in the bulk damage threshold, and reduced the volume density of damage centers relative to the density observed in unbaked crystals. The data is summarized in the table.

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Table Median 1-ns, 1064-nm bulk damage thresholds of KDP crystals tested as received and after baking. All crystals were tested by irradiating each site once (1 on 1 tests), and some sites on some crystals were conditioned by subthreshold shots and then repeatedly irradiated until damage occurred (n on 1 tests).

Baking temperature	Baking time	Median Damage Thresholds, J/cm ²	
		<u>1 on 1 Tests</u>	<u>n on 1 Tests</u>
unbaked	--	7	11
110°C	4 days	8	14
140°C	1 and 4 days	8	18
165°C	1 and 4 days	10	15

We measured damage thresholds on two KDP samples at pulse durations ranging from 1 nsec to 20 nsec and found they increase more slowly than $\sqrt{\tau}$. Baking and laser-hardening these samples also resulted in increased damage thresholds at all pulse durations.

1. Introduction

Potassium dihydrogen phosphate (KDP) and its isomorphs are widely used to generate harmonics of laser frequencies and as electro-optic materials. Systems employing these materials range from small commercial lasers and components to large lasers for inertial confinement fusion research. We and others,¹⁻³ have observed that the usable beam fluence in KDP and its isomorphs is limited by the occurrence of laser-induced bulk damage.

We have recently studied the resistance of KDP crystals to laser-induced damage by 1064-nm, 1-20 nsec pulses. We tested ~100 samples from five manufacturers. The bulk damage threshold fluences, shown in figure 1, range from 2 to 18 J/cm² at 1-nsec; the median is 7 J/cm². In all crystals, damage occurred at isolated centers in the bulk and the volume density of damage points was much greater than that of microscopically visible, pre-existing defects.

Although we have not identified the inclusions responsible for the observed bulk damage, we have observed two interesting and probably related effects -- the bulk damage threshold of KDP crystals can be increased by factors ranging from 1.5 to 5 by irradiating them with laser pulses at fluences below the single-shot threshold or by first baking the crystals and then laser irradiating them.

This paper describes the measurements and results of baking and laser hardening of ~30 KDP crystals at 1064-nm and 1-nsec and two KDP crystals at 1064-nm and 1, 3, 6, 9 and 20-nsec.

2. Observing Bulk Damage in KDP

In our measurements we used the laser damage facility at LLNL. A 6-m focal-length lens was used to converge the beam. The sample was placed at a point where the beam diameter (e^{-2} in intensity) was ~2.5 mm. We employed a vidicon camera to record the beam profile, a calorimeter to measure the pulse energy, and a computer to calculate the corresponding peak fluence at the sample position. An F14A biplanar photodiode and Tektronix 519 oscilloscope were used to determine the pulse duration. Our usual procedures for measuring thresholds was to irradiate several

focal volumes once each at progressively higher fluences, until damage was observed. Before and after each laser shot, we examined the irradiated volume using a collinear HeNe laser beam (~ 1 mm FWHM). The presence of preexisting inclusions or induced damage sites was detected by observing small angle forward scattering in the HeNe beam. The number of damage centers observed in the volume inspected (~ 0.01 cm³) ranged from 1 to ~ 100 . The density of damage sites for fluences two and one-half times the threshold fluences is $10^3 - 10^7$ cm⁻³. In most crystals we tested, the observed volume density of damage centers was >100 cm⁻³, and the procedure described above yielded a well-defined threshold fluence. In a few samples, however, site-to-site variations in damage resistance were observed.

3. Laser Hardening

We were concerned that the number and size of damage centers might increase under repeated irradiation and that damage might occur after many sub-threshold laser shots. Because of these concerns, we irradiated several focal volumes with many shots (n-on-1 tests). In general, if damage occurred on the first shot at a given fluence level, neither the size nor the number of damage centers increased upon further irradiation of that focal volume at the same fluence. Also, we never obtained damage by repeated sub-threshold irradiation of a given volume. In fact, we unexpectedly found that repetitive sub-threshold shots hardened the material against damage at fluences higher than the measured single-shot threshold fluence.

To study the effect of sub-threshold irradiation on a particular crystal, we first measured its single-shot threshold as described above. We then applied sub-threshold shots to undamaged focal volumes. Figure 4 demonstrates the increase in damage resistance obtained by low level irradiation of a crystal whose single shot threshold was 3.0 ± 0.3 J/cm² at 1064-nm and 1-nsec. We irradiated five volumes, indicated by arrows in figure 2.

The five volumes were irradiated with laser pulses as follows: volume 1, one shot; volume 2, two shots; volume 3, three shots, etc. The fluences are listed in table 1.

The sample was rotated 90°, and a damaging shot was taken across the pretreated volumes. The hardened volumes are clearly visible as gaps in the dense trail of damage sites.

In figure 3 we illustrate hardening in a second crystal. Each laser shot is represented by a bar that denotes the fluence level ($\pm 10\%$) and a numeral giving the number of damage center per cm³ caused by the shot (one damage center in the observed volume corresponds to approximately 100 cm⁻³). In single-shot tests, no damage occurred below 4.2 J/cm², but damage centers were produced by each shot with fluence exceeding 5.3 J/cm². We then irradiated two sites with a sequence of shots beginning at fluences below the single-shot threshold. On the first of these sites, no damage occurred during five shots at fluence levels up to 10.5 J/cm². A single damage center was produced at 12 J/cm². The focal volume remained unaltered through seven additional shots, and then damaged (~ 50 centers) at 18 J/cm². The second site was undamaged by

six shots at fluence levels up to 12.5 J/cm^2 . On both sites, the damaging fluence was larger than the single-shot threshold, and when damage occurred the number of damage centers was much lower ($\sim 10^{-1}$) than obtained by single-shot irradiation at comparable fluence. The effect appears to be permanent at room temperature; over a period of a month we observed no degradation in the threshold values of hardened volumes.

In table 2, we give a comparison of single-shot (1-on-1) and laser-hardened (n-on-1) thresholds for 7 crystals from four manufacturers. In KDP samples with single-shot thresholds below the median, 7 J/cm^2 at 1-nsec, laser hardening raised the thresholds to above the median. In a few volumes of a few KDP crystals (e.g. sample 7 of table 2) whose single-shot thresholds were above the median, we were unable to improve the damage threshold by laser hardening. In most volumes of these same crystals, however, laser-hardening was successful in raising the threshold.

4. Combined Baking and Laser-Hardening

To investigate the hypothesis that the laser-hardening effect is thermal, we baked crystals at 110°C , 140°C and 165°C . We determined the damage threshold of these crystals, then laser-hardened and again damage tested them. The maximum baking temperature, 165°C , was chosen to be safely below the 180°C temperature at which KDP undergoes an irreversible transition to a different crystalline state. The transition results in internal fracture and a milky appearance.

The results of the measurements are presented as histograms in figure 4. Damage threshold fluences obtained before the crystals were laser hardened are indicated by open squares, and those obtained after hardening by solid squares. The baking temperatures ($\pm 5^{\circ}\text{C}$) are indicated in the upper left hand corner of each histogram. Median values are denoted by an "M".

Figure 4a presents the data of table 3 plus other data. Figure 4b, 4c and 4d are histograms of the damage thresholds of those crystals heated to 110°C , 140°C and 165°C respectively. Included in all of these latter three histograms are thresholds of crystals baked for 24 hours and for 96 hours. Use of different baking times did not produce any obvious difference in the crystals.

Table 3 presents individually the data shown graphically in figures 4b, 4c and 4d. The monotonic increase of the median threshold with baking temperature was the result of slight improvements in some crystals and larger increases in others. For all crystals, laser hardening produced an increase in threshold that was larger than that obtained by baking alone.

We also observed that baking resulted in fewer damage centers at the same multiple above the single-shot threshold and that the damage centers were smaller.

To see if baking caused any deterioration in harmonic generation, we determined the second harmonic generation parameters of a KDP crystal before and after baking at 165°C for 24 hours; we observed no change.

Because KDP has a low decomposition temperature, we suspected that the damage threshold might be temperature sensitive. Therefore, we determined the 1-on-1 damage threshold of a KDP crystal at room temperature, 18°C, and at 85°C. The thresholds were 6.9 J/cm² and 4.7 J/cm², respectively. Extrapolating these two data points indicates decomposition at 220°C, close to the reported decomposition temperature.

5. Pulse Duration Dependence

We tested two KDP samples at 1, 3, 6, 9 and 20-nsec with 1064-nm pulses. One sample (number 2) had an average bulk damage threshold (7.4 J/cm²) at 1-nsec, while the other (number 1) was below average (3.1 J/cm²). Each sample was tested at the five pulse durations before baking or laser-hardening, then laser-hardened and retested. They were then baked at 140±5°C for 24 hours and retested. They were laser-hardened and again damage tested. The data from these tests are presented in figure 5, where a $\sqrt{T}$ curve is compared to each set of data for reference. The upper error bars indicate the lowest fluence at which damage was observed, and the lower error bars the highest fluence at which no damage was seen. In cases where the "damage" and "no damage" fluences were closer than 10% of the average of the two, the error bars are 10% of the average, which is the system fluence measurement accuracy.

The data for the below average crystal is presented in figure 5a. The scatter in the 1-on-1 data is probably due to nonuniformity in the crystal. In the n-on-1 tests, the scatter can be due to both crystal nonuniformity and variations in the laser-hardening sequence.

In figure 5a, the n-on-1 data, before and after baking are within the range of uncertainty of each other. Although the effect of baking on the 1-on-1 tests is definite, laser-hardening efficiently heats whatever would have damaged in the crystal and little or no further improvement by baking is evident in this crystal.

Figure 5b shows the dependence of threshold on pulse duration, baking and laser-hardening for the average-damage-threshold crystal. Even though the original damage threshold of this crystal is higher than the below average crystal, the final "baked and laser-hardened" data are at nearly the same level.

In the eight sets of data presented in figure 5 a straight line fit would have a slope less than one-half in all cases. Since the data is concerned with just two samples and relatively few determinations on each sample, there are few pulse duration dependence conclusions that can be drawn.

6. Discussion

The demonstration that the bulk damage threshold of KDP can be increased by baking and sub-threshold laser irradiation is unambiguous, but the exact mechanism for the increase is unknown. Bulk damage in KDP at fluence of 2 - 20 J/cm² is likely due to extrinsic absorbing inclusions, such as bubbles containing KDP solution, impurities, metallic particles, and misoriented KDP microcrystallites. Unfortunately, the sites at which damage occurs are so small that we have been unable to observe or locate them prior to laser-damage. Identification of the

absorbers has, therefore, not been made, and hypotheses about the hardening or baking process, including such mechanisms as changes in structure, chemistry, or valance of impurities, are speculative at best.

7. Conclusions

The bulk damage threshold of KDP can be increased from 1.5 to 5 times its single-shot threshold fluence by a combination of baking and sub-threshold irradiation. The degree of improvement depends upon the individual crystal. From our data, we recommend the following threatment procedure to produce a more damage resistant KDP component:

- 1) Bake the crystal at 140°C for at least 24 hours (with an ~ 24 hour cool down period to prevent fracture)
- 2) Initially irradiate the crystal at no more than 3 GW/cm^2 at 1-nsec or $.5 \text{ GW/cm}^2$ at 20-nsec.
- 3) Irradiate with fluence increasing $\sim 1.5 \text{ GW/cm}^2$ at 1-nsec or $\sim .25 \text{ GW/cm}^2$ at 20-nsec until a fluence just below the operating fluence is reached.

Although the baking temperature and baking duration do not seem to be critical, the initial fluence and the details of the laser-hardening sequence are. Figure 6 illustrates this point. The sequence on site number 1 was disrupted by an unusually large laser output resulting in a

jump from 13 J/cm^2 to almost 21 J/cm^2 and one damage center. At the second site an evenly spaced sequence resulted in no damage until almost 36 J/cm^2 . Even here, the last two shots in the sequence were separated too widely. Some of the effect seen in this figure may be due to the non-uniformity of the crystal, but in general the details of the laser hardening sequence are important.

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Figure Captions

1. Histograms of the bulk laser-damage threshold fluence of 65 KDP samples from five manufacturers.
2. Demonstration of increased damage threshold resulting from pulsed laser irradiation of KDP.
3. A comparison of 1-on-1 and n-on-1 laser irradiation of KDP, showing the increased damage resistance of multiply-irradiated sites.
4. Histograms of the bulk laser-damage threshold of KDP: a) as grown before and after laser-hardening b) baked at 110°C for 24 hours, c) baked at 140°C for 24 and 96 hours and d) baked at 165°C for 24 and 96 hours.
5. Dependence of the KDP bulk damage threshold fluence on pulse duration: a) sample 1, b) sample 2.
6. The effect of fluence step size in laser-hardening of a KDP crystal; 1064 nm, 20 nsec.

Table 1. Laser-hardening fluence data for the crystal whose damage photo is shown in figure 1.

Shot/Volume	Fluence (J/cm^2) at 1064-nm and 1-nsec				
	1	2	3	4	5
1	1.1 $\pm$ 0.1	1.1 $\pm$ 0.1	0.67 $\pm$ 0.07	1.3 $\pm$ 0.1	1.1 $\pm$ 0.1
2		2.4 $\pm$ 0.2	2.5 $\pm$ 0.2	2.5 $\pm$ 0.2	2.5 $\pm$ 0.2
3			3.0 $\pm$ .3	2.8 $\pm$ 0.3	3.1 $\pm$ 0.3
4				3.9 $\pm$ 0.4	4.9 $\pm$ 0.5
5					5.3 $\pm$ 0.5
Tranverse(6)		7.6 $\pm$ 0.8 J/cm^2			

Table 2. The effects of laser hardening on unbaked KDP samples

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Sample Number	Bulk Damage Threshold (1 nsec, 1064-nm)	
	1 on 1	n on 1
1	$2.9 \pm 1.0 \text{ J/cm}^2$	$9.1 \pm 0.9 \text{ J/cm}^2$
2	4.4 ± 0.9	9.0 ± 0.9
		11.1 ± 1.1
		16.1 ± 2.5
3	7.6 ± 1.4	11.4 ± 1.3
		16.1 ± 1.9
4	7.7 ± 0.7	11.8 ± 1.7
		13.2 ± 2.2
5	8.4 ± 0.8	14.2 ± 2.8
6	9.3 ± 1.2	9.2 ± 1.7
7	9.4 ± 0.9	8.9 ± 0.9
		12.7 ± 1.3
		13.9 ± 1.4

Table 3. The effects of baking and laser hardening on the bulk damage threshold of KDP.

Bulk Damage Threshold (J/cm ² @ 1-ns, 1064-nm)							
Sample Number	Single Shot Threshold	110°C	Laser Hard. 110°C	140V	Laser Hard. 140°C	165°C	Laser Hard. 165°C
1	3.2±0.3	8.0±1.2	14.4±1.7				
2	3.3±0.5	3.8±0.7	10.9±1.1	4.3±0.4	13.7±1.4	7.6±1.3	10.0±1.3
3	5.0±0.5			7.4±0.7		16.2±1.7	25
4	5.2±0.5	5.9±0.6	15.2±1.6	6.9±0.8	10.8±1.3	8.6±1.0	13.3±2.0
5	6.4±1.3			8.7±0.9	17.2±4.1	10.9±1.6	13.9±1.4
6	6.6±0.7			8.6±1.1	19.3±4.4		
7	6.9±1.0			15.1±2.0	20.4±2.0		
8	6.9±0.7					7.2±0.7	13.2±1.3
9	8.5±1.0					11.3±1.1	17.4±1.7
10	8.5±0.8					8.5±0.9	
11	8.7±0.9	13.0±1.3	14.5±1.6	11.7±1.2	21.3±3.7		
12	9.7±1.0	8.1±0.8	14.3±2.6			10.2±1.0	16.5±1.7
13	10.0±1.0					10.3±1.3	19.9±3.3

LASER DAMAGE THRESHOLDS OF LLNL-TESTED KDP SAMPLES

50-50-0581-1424

Figure 1

**DEMONSTRATION OF INCREASED DAMAGE THRESHOLD
RESULTING FROM PULSED LASER IRRADIATION OF KDP**

50-50-0681-1743

Figure 2

LASER IRRADIATION OF A KDP SAMPLE INCREASES ITS RESISTANCE TO LASER DAMAGE

50-50-0581-1421

Figure 3

BULK DAMAGE HISTOGRAMS OF KDP;
1 nsec, 1.064 μm

50-50-0981-2986

Figure 4

DEPENDENCE OF THE KDP BULK DAMAGE THRESHOLD FLUENCE ON PULSE DURATION – SAMPLE 2

50-50-0981-3091A

Figure 5a

DEPENDENCE OF THE KDP BULK DAMAGE THRESHOLD FLUENCE ON PULSE DURATION – SAMPLE 1

50-50-0981-3091

Figure 5b

THE EFFECT OF FLUENCE STEP SIZE IN LASER HARDENING OF A KDP CRYSTAL

50-50-0981-3090

Figure 6