Laser damage to production- and research-grade KDP crystals

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ABSTRACT

We present the results of laser damage measurements conducted on potassium dihydrogen phosphate (KDP) and deuterated potassium dihydrogen phosphate (KD*P) crystals that were grown recently for both production and research applications by several sources. We have measured extrinsic damage thresholds that cover wavelengths from 1064 nm to 266 nm at pulse durations in the 3- to 10-ns regime.

Many of the samples were extracted from boules grown specifically to yield large-area crystals, up to 32-cm square, for laser fusion applications. These crystals were the result of efforts, both by the Lawrence Livermore National Laboratory (LLNL) and commercial crystal-growth companies, to yield high-threshold KDP. In particular we have established that such crystals can reliably survive fluences exceeding 15 J/cm^2 at 355 nm and 20 J/cm^2 at 1064 nm when irradiated with 3-ns pulses. We present details of how bulk and surface damage to these crystals scale with pulse duration and wavelength as well as of morphological effects due to laser conditioning.

1. INTRODUCTION

In 1991 we established conservative guidelines for the bulk damage thresholds of KDP crystals for use in large-aperture laser systems such as LLNL's existing Nova laser, planned Beamlet laser, and proposed National Ignition Facility (NIF). They were based on our experience of laser-induced damage measurements that we have conducted on crystals grown both at LLNL and by commercial and research institutions in a collaborative effort with LLNL.[1,2] Our empirical assessment at that time indicated that, with pulse durations scaled to 3-ns, we could expect to achieve damage thresholds of 24 J/cm^2 at 1064 nm (1ω), 20 J/cm^2 at 532 nm (2ω), 11 J/cm^2 at 355 nm (3ω), and 3 J/cm^2 at 266 nm (4ω). We had in fact reported higher thresholds but allowed for conservative "derating" of thresholds for full-sized, production optics.[3]

This year we conducted a series of damage measurements at 1ω and 3ω on 27 KDP and KD*P crystals that were for the most part production-grade crystals with nominal dimensions of 50 x 50 x 10 mm. Most of these came from boules that were grown by commercial vendors or research institutions to produce optical-grade crystals with dimensions as large as 32-cm square. Our current tests included such diverse sources as Cleveland Crystals, Inc., Inrad, Optochemical Corp., and Moscow State University. The surfaces were prepared either by the respective crystal growers LLNL or Canon. Over a period of 15 years we have tested crystals from 13 different sources. However, for proprietary reasons we cannot ascribe specific vendor names to the test samples under discussion here. Not all crystals had acceptable bulk and surface characteristics for use in a large-aperture laser system. Those KDP crystals that did had bulk thresholds that ranged from 24 to 34 J/cm^2 at 1ω and 15 to 20 J/cm^2 at 3ω when measured with 3-ns pulses. The KD*P crystals had average thresholds that were essentially comparable to those of KDP at 3ω. At 1ω they were approximately 20% lower. In Fig. 1 we show these ranges of bulk thresholds superimposed on a plot of our earlier guidelines. Most current production KDP crystals have exceeded these guidelines.

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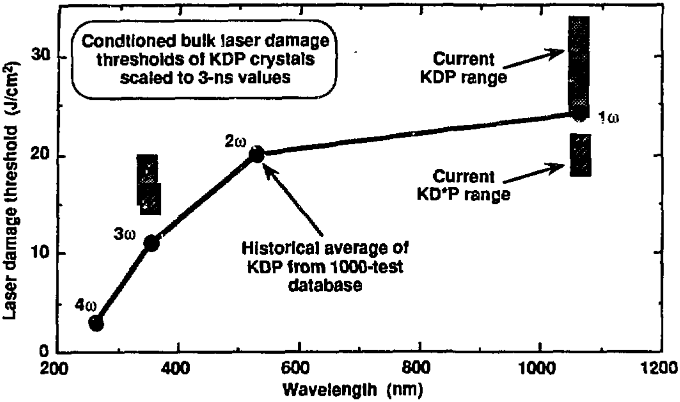

Fig. 1. Ranges of the conditioned, bulk, laser-damage thresholds of the current series of acceptable KDP and KD*P crystals measured with 3-ns pulses at 1ω and 3ω. These are superimposed on a plot of previous thresholds based on over 1000 damage measurements to KDP crystals at 1ω, 2ω, 3ω and 4ω.

2. EXPERIMENTAL PARAMETERS

We conducted all of the current laser damage measurements with the Chameleon laser facility at LLNL.[4] This system was recently upgraded so that we could conduct these measurements not only at the two critical harmonics but, more importantly, at two pulse durations. These durations cover both a major portion of our earlier KDP damage database at 10 ns and our immediate requirements at 3 ns. The pertinent parameters for this system are enumerated in Table 1.

Wavelengths:	1064 nm (1ω) and 355 nm (3ω)
Pulse durations:	3 ns and 10 ns full-width half-maximum
Irradiation rate:	10 Hz for 1 minute
Beam size:	0.9 - 1.9 mm diameter ($1/e^2$) smooth gaussian
Number of pulses per site:	Up to 600 unless massive damage occurred
Irradiation types:	Unconditioned (S/1) — 600 shots of ~ equal fluence Conditioned (R/1) — 600 shots ramped up in fluence
Peak fluences:	For 1ω ~ 80 J/cm^2 @ 10 ns, ~ 40 J/cm^2 @ 3 ns For 3ω ~ 40 J/cm^2 @ 10 ns, ~ 25 J/cm^2 @ 3 ns
Nominal threshold error	± 15%

Table 1. Parameters for laser damage testing of current KDP and KD*P crystals

In Fig. 2 we show a schematic of the irradiation and damage detection mechanism. Our samples were mounted in our standard microscope stage. This allowed us to (1) translate the samples through the test beam in the irradiation position, and (2) check them for damage both before and after irradiation by swinging them into the examination position. Rather than conventional Nomarski illumination, we employed a fixed fiber-optic light source to illuminate the sample from the rear. The light was mounted so that damage appeared as bright, scattered, white light in a dark field. This proved to be more sensitive to damage detection than Nomarski microscopy. In all cases, we examined for and specified damage in three locations: (1) the incident or ***front*** surface, (2) the exit or ***rear*** surface, and (3) the ***bulk*** material. The latter two were accomplished by zooming through the bulk material with sufficient working distance to range through the full 10-mm thickness of each crystal. In some instances we also examined the rear surfaces for post-irradiation damage by reversing the sample in the microscope since this provided better resolution. Damage determinations were made based on visual and photographic assessments at 100x magnification.

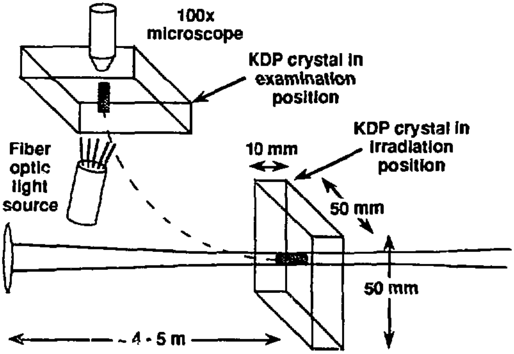

Fig. 2. Experimental configuration for irradiating and examining a 1-mm-diameter x 10-mm-thick cylindrical volume of a KDP crystal.

Depending both on which wavelength and pulse duration employed, we irradiated the samples at the beam waist of a smooth Gaussian beam profile focused from a distance of 4 to 5 m. The waist had beam diameters of 0.9 to 1.9 mm which remained effectively fixed through the 10-mm thickness of the crystals. Such "large-spot" measurements allowed us to irradiate relatively large volumes of the crystals at high fluences to locate isolated damage points. This contrasts with "small-spot" measurements which focus a "hot" beam into a crystal volume up to 10,000 times smaller. The latter can yield high intrinsic damage thresholds but fail to locate the isolated, damage-causing defects which limit a crystal's operating fluence range in a full-sized laser system. The spot sizes were governed by the available energies for each parameter condition so that we could achieve sufficient fluence to induce damage in the bulk material. Hence, we had to use smaller spots at 3ω than at 1ω and also for 3-ns pulses than for 10-ns pulses. Since we were able to examine a cross section of about 1 mm^2 without translating the sample, we therefore searched for bulk damage through a volume of about 10 mm^2 per site.

3. DAMAGE THRESHOLDS AND MORPHOLOGIES

3.1 Bulk damage at 1ω and 3ω with 3-ns pulses

Our primary objective was to determine the survivability of these crystals under our large-system operating constraints which would employ pulse durations of 3 ns at 1ω and 3ω. In Fig. 3 we summarize the results for all of our measurements on 19 KDP crystals and 8 KD*P crystals which were irradiated with 600 shots using conditioning irradiation. The conditioning process is explained below. We have ranked the samples by decreasing 3ω thresholds for KDP and separately for KD*P. The technical thresholds, based on the minimum observable damage, are shown by the heights of the black columns. The 1ω thresholds fall directly below the corresponding 3ω values. This same ranking order will be employed to compare other damage parameters in subsequent figures. The shaded regions in the background define the aforementioned threshold regions of crystals that had "cosmetically clean" surface and bulk regions and also high thresholds. These values are in fact greater than those required by a NIF laser.

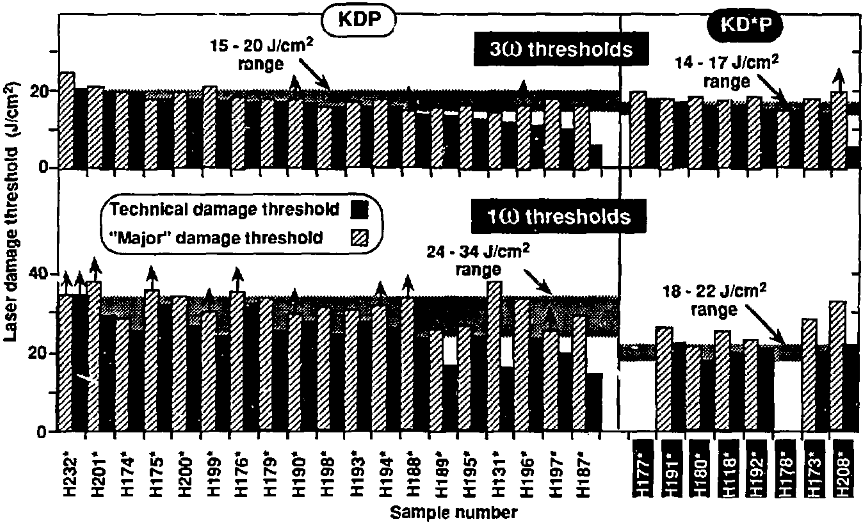

Fig. 3. Conditioned bulk laser damage thresholds of 27 KDP and KD*P crystals measured with 3-ns pulses. The samples are ranked in order of decreasing 3ω thresholds for each crystal type. The black columns indicate technical thresholds whereas the shaded ones show thresholds for major damage. The shaded bars give the threshold ranges of good quality crystals.

The damage thresholds for "major" bulk damage is shown in the adjacent shaded columns. By our strict assessment of bulk damage, we determine that a crystal has been damaged even if we can detect only one bulk pinpoint within a detection limit of 1 to 10 μm in the 10-mm^3, examined volume. If such small damage does not continue to grow it would have no effect on the operation of an actual large laser since its effects would quickly diffract out. In fact, although > 99% of all test sites that we examined in the current series of crystals were free of pre-existing defects of such dimensions, occasionally we still did run across isolated, pre-irradiation defects ≤ 10 µm in size. For the current tests we have arbitrarily established a "major" bulk damage threshold where we either observed ≥ 10 pinpoints of new bulk damage ≤ 10 µm in size, or at least one damage cluster or fracture ≥ 30 µm in size. We found that at 3ω the major bulk damage threshold averaged 19% higher than our strict technical threshold. At 1ω it was 37% higher. An arrow in Fig. 3, and in corresponding subsequent figures, means that the threshold for that particular sample was higher (or lower) than the irradiation fluence shown.

We observed that the damage morphology often consisted of tiny pinpoints ≤ 10 µm in size. At 100x magnification we were not able to resolve this damage in great detail. At higher magnifications we lacked sufficient working distance with our objective lenses to zoom far enough into the bulk material to be able to detect damage. However, at higher fluences we were able to observe the damage morphology change in four ways: (1) the density of fine pinpoints grew so that they could often be detected by naked eye as a "smoke trail" when illuminated with bright light; (2) the micro explosions which generated the pinpoints got more massive yielding larger, jagged clusters 20 to 200 µm in size; (3) these clusters developed single or double, orthogonal fractures which ranged up to 400 µm in length and were always oriented in the same directions; and (4) the crystal failed massively, usually necessitating that we had to cease irradiation. In Fig. 4 we show examples of (1) and (3) at 100x magnification with back-lit, dark-field microscopy. The left picture is focused on a plane within the bulk of a "smoke trail" showing many pinpoints out of focus in other focal planes. The right picture shows a typical 300-µm fracture through a 100-µm cluster.

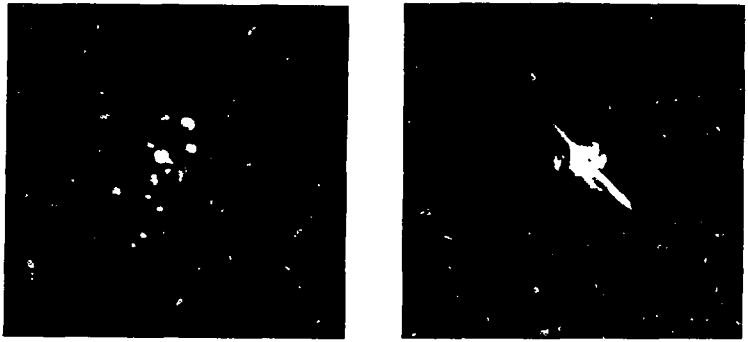

Fig. 4. Typical major bulk damage morphologies. At the left, pinpoints of damage, usually ≤ 10µm in size, degenerated to a"smoke trail" through the bulk material. At the right, a large damage cluster formed with one or two fractures up to 300 µm in length.

3.2 Laser conditioning

We have consistently established that we can increase the bulk damage thresholds of KDP crystals by first subjecting them to conditioning irradiation.[1, 2] This requires that the material be subjected to a series of gradually increasing fluence levels beginning below unconditioned threshold levels. This can be accomplished by irradiating either small sites, one site at a time, during the test sequence, or the entire volume with a full-sized beam using at least six incrementally increasing steps. Since we conducted all of our damage tests for one minute at 10 Hz, we utilized far more shots than necessary to condition our damage test samples. In Fig. 5 we show the irradiation schemes for 600 shots with (1) unconditioned irradiation using nominally the ***same*** fluence per shot (S/1), and (2) with conditioned irradiation (R/1) where the fluence was gradually ***ramped*** up from zero to a desired level. The ramp was accomplished in several hundred shots and the fluence was then maintained at that desired level for the remainder of the 600 shots.

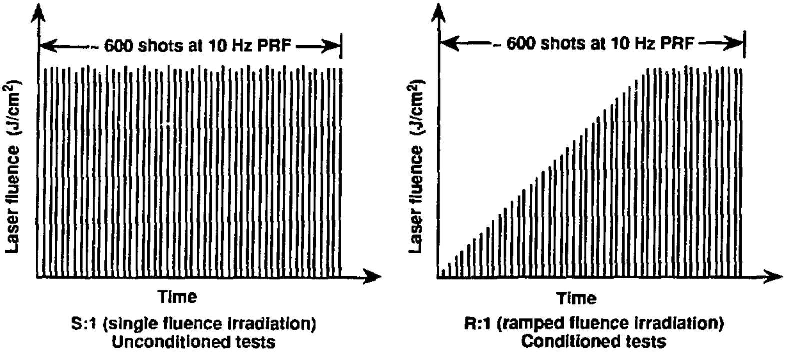

Fig. 5. Unconditioned (S/1) and conditioned (R/1) laser irradiations with 600 shots.

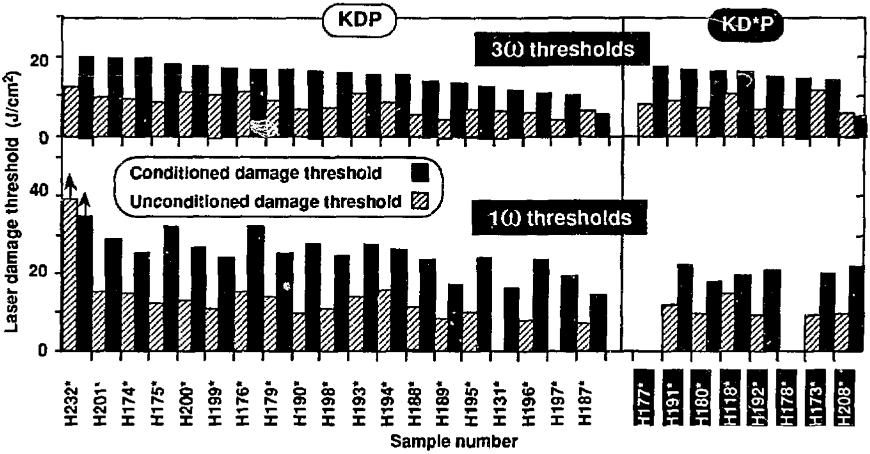

Fig. 6. The black columns show that with conditioning the average damage thresholds at each harmonic were raised by a factor of 2.1 over those irradiated with unconditioned pulses (shaded columns).

The effects of conditioning the bulk material are multifold: (1) the results are permanent, (2) the increase in bulk threshold goes up significantly, and (3) the damage morphology is much more benign at fluence levels above the newly conditioned thresholds. Fig. 6 shows the same ranking of conditioned, 3-ns damage thresholds at 1ω and 3ω in black. The adjacent shaded columns show the corresponding damage thresholds when the samples were measured with unconditioned, S/1 irradiation. We found that for these tests, conditioning raised the damage thresholds by an average factor of 2.1 both at 1ω and 3ω. Conditioning of the full volume of a crystal requires either multistep rastering of the entire volume with a small beam, or multiple shots with a full-sized beam. In our damage measurements we conditioned only the small volume that we irradiated for damage tests. We have therefore not subsequently returned to that same small volume to determine whether a 1ω-conditioned volume would also have been conditioned for use at 3ω or vice versa.

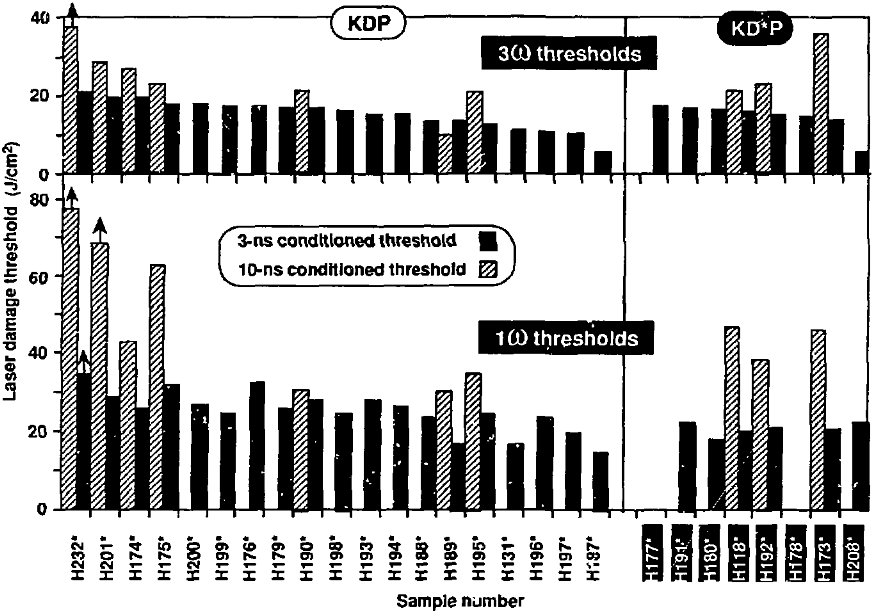

Fig. 7. For the ten samples that were also tested with 10-ns pulses, the pulse durations scaled by an average of $\tau^{0.5}$ from the 3-ns values.

3.3 Pulse-duration scaling

Over a period of 15 years we have conducted more than 1000 damage measurements on KDP crystals. These had typically been conducted with pulse durations ranging from < 1 ns to > 50 ns. Until this year our most recent tests were usually conducted with 10-ns pulses. We would like to be able to scale many of these data to a relevant pulse duration τ, in our case 3 ns. Past empirical results have led us to a KDP bulk scaling relationship of $\tau^{0.5}$ at both 1ω and 3ω. This turns out to be the same conventional rule-of-thumb based on thermal diffusion most people use in scaling laser damage data. However, in reality most optics usually scale at a lower rate. We conducted comparable damage measurements with 10-ns pulses at both 1ω and 3ω on 10 of the 27 samples in this study. The conditioned results are compared with the corresponding 3-ns thresholds in Fig. 7. For these recent KDP samples we found that the scaling factors averaged at > $\tau^{0.43}$ at 1ω and > $\tau^{0.24}$ at 3ω; the corresponding KD*P scaling factors were $\tau^{0.63}$ and $\tau^{0.43}$ respectively.

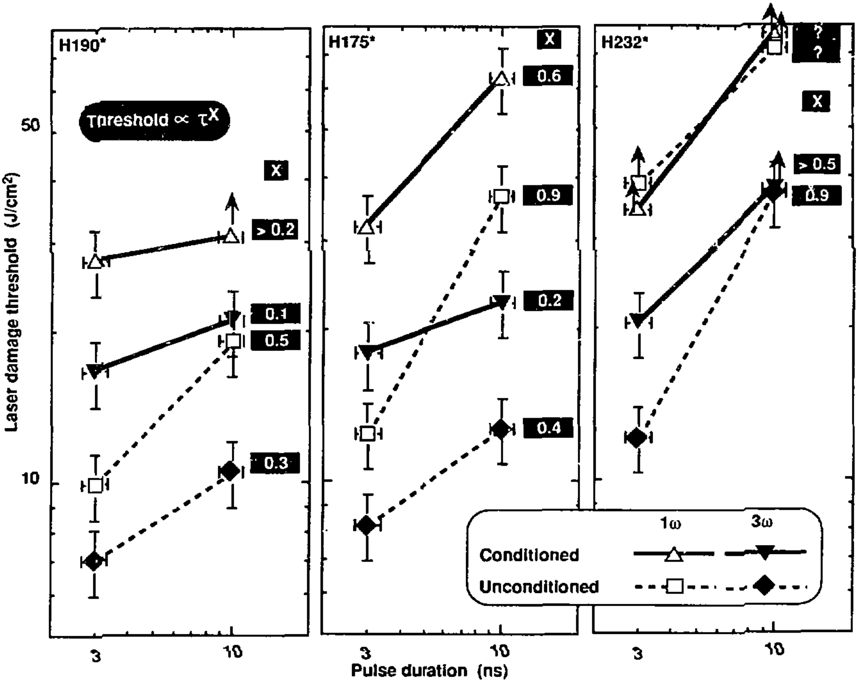

Fig. 8. Low-, moderate-, and high-threshold crystals appeared to have pulse duration scaling factors that ranged over a very wide range in spite of moderate measurement error bars. However, the average factor of $\tau^{0.5}$ for all tests had a large error range of $\tau^{+0.4/-0.3}$ because the spread in pulse durations of 3 to 10 ns was actually relatively small.

In Fig. 8 we show the conditioned and unconditioned results plotted on log-log scales for representative low-, moderate-, and high-threshold KDP crystals. These display scaling factors ranging from $\tau^{0.1}$ to $\tau^{0.9}$. Since we were unable to induce 1ω bulk damage in the best crystal, those scaling factors are shown as indeterminate. The variation in factors implies that a note of caution be observed regarding the credibility of scaling factors with relatively small spreads in pulse durations (3 and 10 ns in our case). Our ± 15% error bars in measured damage thresholds and ± 10% error bars in measured pulse durations are not particularly large. Yet the resulting errors in a $\tau^{0.5}$

scaling factor for such measurement errors would lead to scaling errors of $\tau^{+0.4/-0.3}$. Therefore, our average values, based on only a few sets of measurements, are in relatively good agreement. More definitive measurements would require that damage measurements be conducted at more widely spread pulse durations. Comparable measurement errors for tests with 1- and 20-ns pulses, for instance, would reduce the scaling errors of a $\tau^{0.5}$ scaling to $\tau^{+0.17/-0.14}$.

3.4 Wavelength scaling

It would be convenient to be able to generate a comparable scaling relationship for the bulk damage thresholds of KDP crystals with wavelength. The average data points of our historical summary from over 1000 measurements (Fig. 1) don't readily lend themselves to any simple scaling law. We roughly fit these scaled, 3-ns data to the following empirical wavelength scaling relationship:

Threshold $\approx$ 36.7 (log λ) - 83.1, (threshold in J/cm^2 for 3-ns pulses and λ in nm).

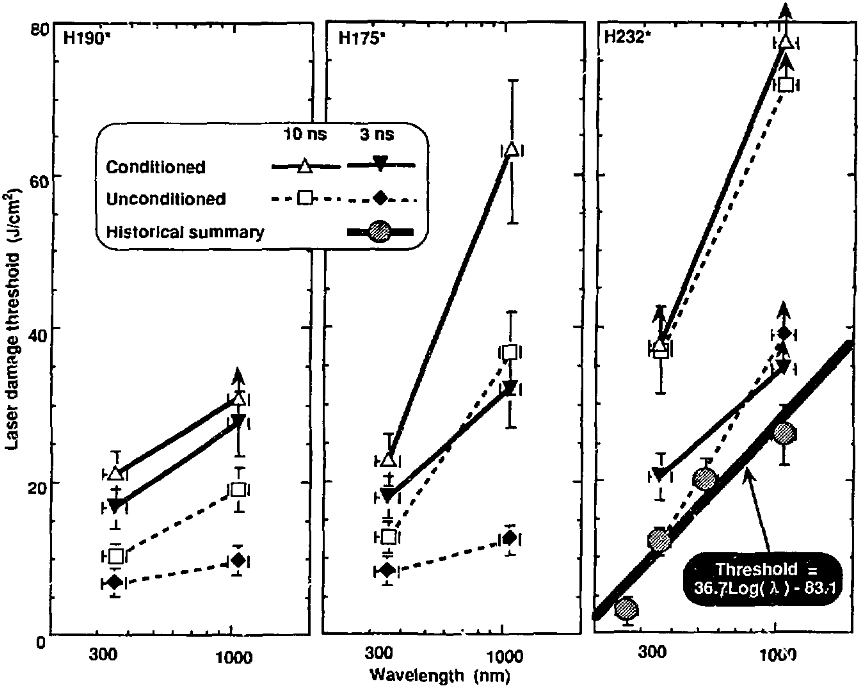

Fig. 9. Conditioned damage thresholds with 3-ns pulses scaled roughly like an empirical scaling approximation generated from our database of 1000 KDP bulk damage tests.

Fig. 9 shows how the same three samples of KDP crystals used in Fig. 8 would compare with this wavelength scaling relationship for 3- and 10-ns pulses using conditioned and unconditioned irradiation. To first order there was at least moderate agreement with the 3-ns data considering that (1) for the current results we have only two wavelength data points, and (2) our current samples have improved in damage thresholds over those from our historical averages.

3.5 Surface damage thresholds

We have concerned ourselves primarily with bulk damage thresholds. However, for each measurement, we also attempted to assess the effects of damage to the bare, uncoated surfaces. The surface preparations fell primarily into two categories: (1) those prepared by diamond turning, and (2) those prepared by lap polishing. Even within these categories there were different vendors employing different methods with varying results. Not all of these were necessarily documented. For proprietary reasons we again do not specify which samples had been figured by which process or vendor. In general, we found that the diamond-turned surfaces exhibited superior damage resistance and cosmetic appearances over their polished counterparts.

All of the samples had been cleaned at LLNL by our standard procedures used on the Nova laser system. These included a three-step process in a clean-room environment involving: (1) a 24-hour bath in toluene, (2) followed by a 90-psi toluene spray of the vertically mounted surfaces until uniform sheeting was observed, (3) concluded with a toluene drag wipe. The samples were then transported in individually sealed containers until they were damage tested in a filtered-air clean hood.

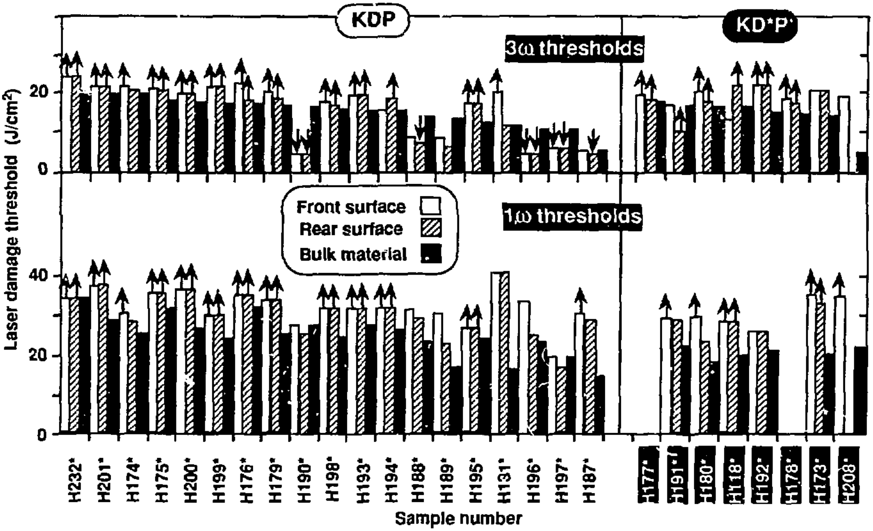

Fig. 10. Surface damage thresholds generally exceeded those of the corresponding bulk material. Diamond-turned surfaces yielded little or no damage at either wavelength. Polished surfaces exhibited significant highlighting of polishing sleeks and pits at 3ω.

Fig. 10 shows the results of these surface damage assessments compared to the previously noted bulk thresholds using conditioned irradiation. For this figure the white columns represent front-surface thresholds and the shaded columns rear-surfaces ones. In most instances we found that the surfaces had thresholds that comfortably exceeded the bulk damage thresholds. This was particularly true for the diamond-turned surfaces. Rather than risking massive damage to the bulk material, we often did not proceed with measurements at higher fluences just to firmly establish surface threshold values.

Most of those surfaces that did exhibit definitive damage were ones that had been polished rather than diamond turned. Fig. 11 shows two photographs of a representative polished surface. The left picture (displayed at ~ 80x magnification) shows a high density of polishing sleeks and pits. The largest of the pits were about 20 μm in size. When damage was observed at 1ω it consisted typically of some further new pits and minor enhancements of existing defects. However, at 3ω most of the pre-existing defects were dramatically highlighted. This is shown in the right photograph of the same site after laser irradiation. To the naked eye this generally appeared as a general fogginess to the surface. The busy nature of these surfaces complicated both surface and bulk damage assessments. Many surface thresholds were vague since it was difficult to determine if any new defects had appeared. Scattering from these surfaces also made it difficult to detect subtle bulk damage. In some instances obvious surface defects were the source of massive surface damage which may then have propagated into the bulk material. This often spalled off large pieces of the crystals. Another major surface phenomenon was the occasional emission of bright light from a plasma-generated scald. Although we do not believe that conditioning has the same effect on surfaces as it does to the bulk material, we typically found that the gentle ramping-up of fluence may have cleaned some residual surface contaminants so that "conditioned" surface thresholds were somewhat higher than corresponding unconditioned thresholds. Most of the diamond-turned surfaces wound up being so clean that we typically could not detect any pre- or post-irradiation damage. It was in fact often difficult to know when the microscope was actually focused on the front or rear surfaces of such crystals.

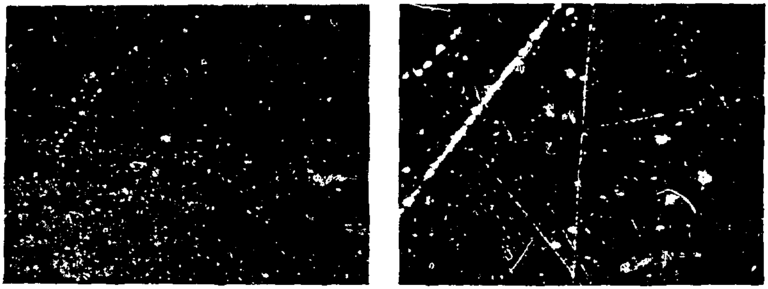

Fig. 11. Polished surfaces at ~ 80x magnification. Pits and sleeks were brightly highlighted when irradiated at 3ω.

4. CONCLUSIONS

We have determined, through the collaborative efforts between LLNL and several commercial vendors of KDP crystals, that these vendors can grow such crystals of sufficient size and with acceptable damage thresholds. Our recent tests were conducted on relatively large crystals with cross sections 5-cm square. Some of these boules were grown for LLNL to yield crystals with cross sections as large as 32-cm square.

The current design for the Beamlet laser now under construction at LLNL imposes peak fluence levels on KDP crystals of 17 and 12 J/cm^2 for 3-ns pulses at 1ω and 3ω respectively. Of those crystals that were cosmetically acceptable we have measured relevant bulk damage thresholds that ranged from 18 to > 34 J/cm^2 at 1ω and from 14 to 20 J/cm^2 at 3ω. These goals were achieved by conditioning the samples with gradually increasing laser fluences to more than twice their unconditioned thresholds. Surface damage thresholds have in general not imposed any restrictions on these operating parameters at 1ω. However, in general, at 3ω only diamond-turned surfaces survived fluences to the same levels as those of the respective bulk materials. We are pursuing efforts both at LLNL and with our vendors to improve the polishing processes so that these surface thresholds at 3ω can be raised to higher levels.

5. ACKNOWLEDGMENT

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