Characteristics of Laser-Induced Surface and Bulk Damage of Large-Aperture Deuterated Potassium Dihydrogen Phosphate at 351 nm *

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Deuterated potassium dihydrogen phosphate damage performance at 351 nm is studied on a large-aperture laser system. Bulk and rear-surface damage are initiated under the 3ω fluences of 6.7 J/cm^2 and 3 J/cm^2, and show different growth characteristics under multiple laser irradiations with the fluence of 6 J/cm^2. The size and number of bulk damage keep unchanged once initiated. However, surface damage size also does not grow, while surface damage number increases linearly with laser shots. Different damage thresholds and growth behaviors suggest different formations of bulk and surface damage precursors. The cause of surface damage is supposed to be near-surface absorbing particles buried under the sol-gel coating.

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Potassium dihydrogen phosphate (KDP) and its deuterated isomorphs (DKDP) are currently the only suitable nonlinear crystals which can be grown to the sufficiently large size needed for frequency conversion in large-aperture high-power laser facilities. In common configurations, a KDP doubler converts the laser wavelength from 1053 nm (1ω) to 526 nm (2ω) and a DKDP tripler mixes the generated 526-nm laser light with the residual 1053-nm light to produce 351-nm light (3ω).[1] DKDP rather than KDP is used as the tripler to suppress transverse stimulated Raman scattering (TSRS) and avoid TSRS-induced damage.[2,3] However, DKDP is more expensive and more vulnerable to laser-induced damage than KDP,[4−6] thus laser-induced damage of DKDP becomes a crucial problem and is regarded as an important factor limiting the high-fluence output of high-power laser facilities.

Numerous research studies have been performed to study the damage characteristics of DKDP at 351 nm.[4−12] It was reported that the DKDP surface damage threshold is significantly higher than the bulk damage, which is the primary limitation of the DKDP performance. Bulk damage typically manifests itself as localized microcavities, commonly referred to as pinpoints.[7] The common recognition is that the main reason for bulk damage is the presence of intrinsic lattice defects and/or impurities generated and/or introduced during the DKDP growth process.[8−11] Thus great attention is being paid to the development of the DKDP growth process, to inhibit the formation of defects and impurities.

However, most damage experiments are conducted off-line with small-aperture beams on small crystal samples. Although such measurements are capable of producing excellent qualitative results, small beams are not fully representative of the power laser beam in terms of size, contrast, peak intensities and phase modulations.[12] For instance, the absolute laser-induced damage thresholds tend to be higher than those found on large-aperture laser systems.[13] In addition, offline damage tests are commonly performed at a single wavelength of 351 nm or 355 nm, while in large-aperture laser facilities, DKDP's are used as the triplers and are irradiated with 1ω, 2ω and 3ω light simultaneously. The residual 1ω and 2ω light will affect the damage performance of the crystals.[14,15] For these reasons, we perform an experiment on a large-aperture laser system to investigate the damage performance of DKDP. It is found that the DKDP bears severe rear surface damage with a much lower damage threshold than the bulk. Moreover, the surface damage growth characteristics are markedly different from the bulk damage. We present the experimental results and give a discussion in this work.

The experiment was carried out on a large-aperture high-power laser system called an integrated test bed which can deliver an output energy of about 15 kJ at the wavelength of 1053 nm with a 0.3 nm bandwidth and a 36 cm × 36 cm beam size.[16] The temporal profile of the laser pulse is flat in time with a pulse duration of 5 ns. The experimental layout is shown in Fig. 1. By passing through an 11-mm-thick type-I KDP doubler and then a 9-mm-thick type-II DKDP tripler, the wavelength of output linear-polarized 1ω beam changes from 1053 nm to 351 nm. A large-aperture sample mirror with an uncoated front surface and an antireflection coated back surface is placed behind the DKDP crystal. It reflects a portion of 3ω light into the precision diagnostic system for the

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energy, beam near field and time profile measurement. An absorbing glass behind the sampling mirror is used to absorb the residual 1ω, 2ω and 3ω light simultaneously.

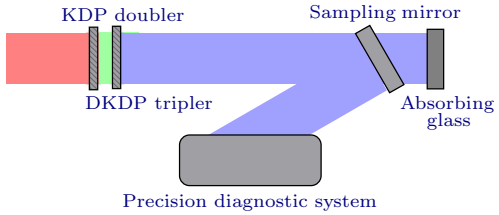

Fig. 1. Schematic diagram of the experimental bench used for DKDP damage tests.

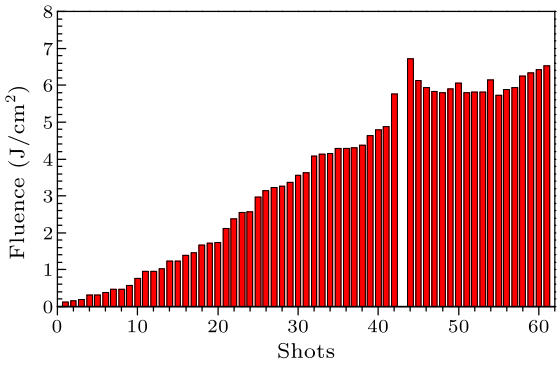

Fig. 2. Fluence of laser shots in the experiment.

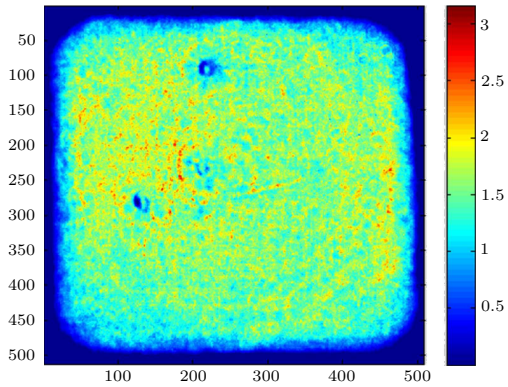

Fig. 3. Typical beam near field of 3ω beam. The modulation ratio and the contrast ratio are 1.77 and 0.13, respectively.

Damage performance of the DKDP tripler is of particular concern in this study. To study its damage characteristic, we perform a damage experiment which contains 60 laser shots that can be divided into two phases, as shown in Fig. 2. The first phase concerns the laser conditioning and damage initiation shots. At this step, 43 shots are fired with the ramped 3ω beam fluence increasing from 0.1 J/cm^2 to 6.7 J/cm^2. It is believed that the laser irradiation of ramping energy could eliminate the possible super-low-damage-threshold defects and therefore could improve damage threshold.[17–19] At the end of the first phase, DKDP bulk damage is initiated at the 43rd laser shot with the highest 3ω beam fluence of 6.7 J/cm^2. Next, 17 additional shots with nearly constant 3ω fluence of 6 J/cm^2 are fired for damage growth testing in the second phase. During this phase, the THG efficiency was about 70%, thus we could reasonably assume that the DKDP mainly bears the 3ω light. As shown in Fig. 3, the 3ω beam near field modulation is below 1.8 in the experiment.

During the experiment, the DKDP crystal is monitored with an online optics damage inspection instrument which is a 400-mm focal length optical telescope with a science-grade CCD camera that can be automatically moved into the beamline. The optical resolution of the online inspection instrument is about 50 μm. The *in situ* pictures of the crystal are taken with this instrument after each laser shot, then the status of the crystal is checked. Once damage occurs, this instrument can provide the information about the size and location of each damage site, thus we could investigate the DKDP damage evolution based on these data.

Fig. 4. Dark-field image of the DKDP crystal after the experiment. The size of DKDP is 430 mm×430 mm×9 mm.

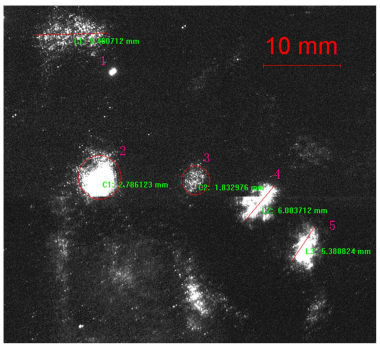

Fig. 5. A cutout from an *in situ* image showing bulk damage sites.

After the experiment, the DKDP crystal is removed from the experimental platform and is examined in a dark room. The dark-field picture of the crystal is given in Fig. 4. It shows bulk damage and severe rear-surface damage including craters and scratches. According to the shot-by-shot information provided by the online optics damage inspection instrument, bulk damage is generated at the 3ω fluence of 6.7 J/cm^2 (the 43rd shot) while the surface damage is initiated under a much lower fluence of approximately 3 J/cm^2.

Bulk damage sites with a typical size of 1–5 mm are shown in Fig. 5. Each damage site is composed of numerous pinpoints. Viewed from the inside, most damage sites are located in the latter part of the crystal and the size of the tail of damage volumes is larger than the head. The reason is that the pinpoint density is proportional to the 3ω fluence which increases from the input surface to the output surface of the DKDP tripler. Thus the size of bulk damage increases along the direction of propagation of the 3ω beam. This damage feature cannot be observed at small-aperture laser systems on which the damage tests are conducted under a fixed fluence along the beam propagation direction.

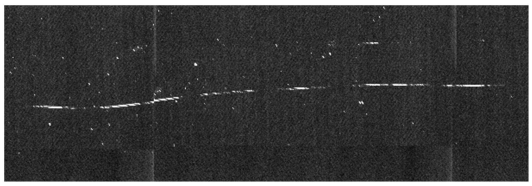

Fig. 6. Scratch on the rear-surface of the DKDP. Its total length is 141 mm.

The phenomenon that the surface damage threshold is lower than the bulk is different from the experimental results obtained at small-aperture laser systems. This is mainly due to the aperture effect, which means that with a small-aperture laser it is harder to detect the dispersed defects and furthermore, a smaller damage testing area will lead to a higher damage threshold. It is commonly known that surface damage of wide-gap materials results from extrinsic nanoabsorbers while bulk damage is caused by intrinsic defects. Extrinsic absorbers produced during both grinding and polishing processes distribute more randomly than intrinsic defects generated in the crystal growth. The aperture effect of surface damage is more obvious than that of bulk damage. This viewpoint is supported by comparing the bulk and surface damage thresholds tested on large-aperture laser facilities to those on small-aperture laser systems. It was reported that the DKDP R-on-1 bulk and surface damage thresholds on a small-aperture laser are about $8\,\mathrm{J/cm^2}$ and $19\,\mathrm{J/cm^2}$ respectively.[4,5] Obviously, the bulk damage threshold at the small-aperture laser system is nearly the same as our results of $6.7\,\mathrm{J/cm^2}$, while the surface damage threshold is about six times higher than ours. It is noteworthy that both the crystals tested in our experiment and in Refs. [4,5] come from the same vendor. It has to be pointed out that the damage fluences $6.7\,\mathrm{J/cm^2}$ and $3\,\mathrm{J/cm^2}$ in our experiment are the beam average fluence, and the peak fluence across the beam is 1.77 times the average fluence.

Another factor that needs to be taken into account is laser conditioning,[17–19] which was employed in the experiment by pre-exposure to increase laser fluences. Laser conditioning is commonly admitted as an efficient process to improve the bulk damage threshold of DKDP crystals due to the fact that it can mitigate the potential defects in the bulk by decreasing their size and/or absorption. Compared with the intrinsic defects, laser conditioning has less effect on the surface damage precursors.

In addition to the bulk damage, there are a large number of damage craters and several scratches on the DKDP rear surface. The length of the longest scratch is 141 mm, shown in Fig. 6. The width of the scratch is about 35 μm and remains unchanged once initiated. According to the *in situ* pictures, these scratches are invisible at the beginning of the experiment and formed gradually under subsequent laser shots. The morphology of scratches strongly suggests that they are not produced during diamond tool finishing of the crystals. It is found that they were produced in the cleaning process prior to the application of the antireflection coating. After sol-gel coating, these scratches are buried under the coating. Under the 3ω irradiations, scratches are opened and weak points along the scratches are damaged. Hence it is necessary to improve the cleaning process of crystals to avoid the introduction of scratches.

Damage growth is our major concern due to the fact that it ultimately limits the lifetime of crystals. After the 43rd shot, 17 additional shots are continuously fired to investigate the damage growth behavior of DKDP surface and bulk damage in the second phase of the experiment. The 3ω beam fluence is fixed at approximately $6\,\mathrm{J/cm^2}$. It is found that no new bulk damage appears and the sizes of pre-existing bulk damage remain unchanged in the remaining 17 shots. The results indicate that not only the bulk damage initiation but also the bulk damage growth of DKDP bulk damage is a threshold event, and the bulk damage is stable below the 3ω fluence of $6.7\,\mathrm{J/cm^2}$.

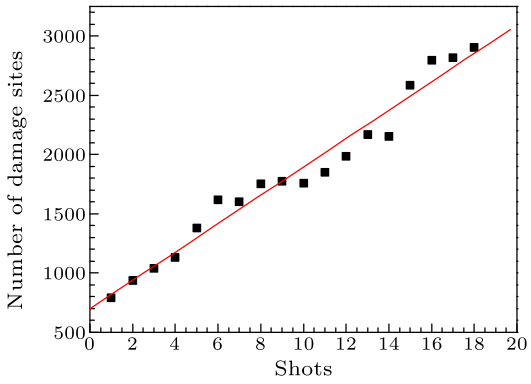

Fig. 7. The number of surface damage sites increases linearly with laser shots at a constant 3ω fluence of $6\,\mathrm{J/cm^2}$.

On the contrary, the surface damage shows a different growth behavior. We use the *in situ* pictures to count the damage sites after each shot, and plot the number of damage sites versus laser shots in Fig. 7. It can be seen that the number of surface damage sites increases linearly with laser shots at a constant 3ω fluence of $6\,\mathrm{J/cm^2}$. This can be explained by the accumulation effect of the beam fluence distribution.[20] As we know, it is inevitable that there are hot spots in

the beam. Although the beam mean fluence is fixed at 6 J/cm^2, the movement of hot spots from shot to shot can substantially increase the number of damage sites, resulting in a growing number of initiations with increasing shots.

We choose twelve separately discernible sites and examine their size-growth behavior site by site. The diameter of each damage site is measured on the *in situ* pictures of the crystal after each laser shot. Figure 8 shows the growth of two individual damage sites with laser shots. As can be seen, both small (with the diameter of 100 μm) and large (with the diameter of 300 μm) damage sites are relatively fixed for tens of shots. This means that the size of surface damage does not grow below the 3ω fluence of 6.7 J/cm^2.

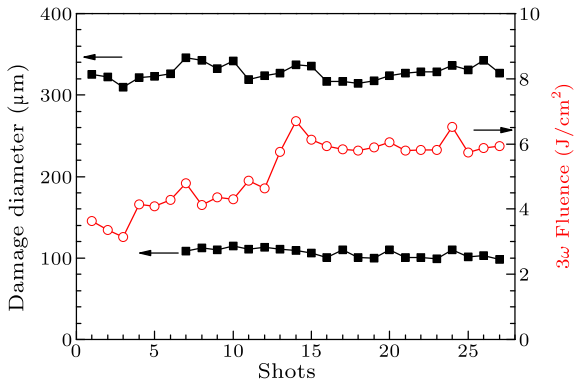

Fig. 8. The size of surface damage sites versus laser shots under different 3ω fluences.

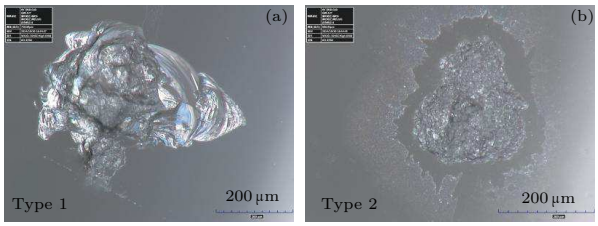

Fig. 9. Typical morphologies of rear surface damage sites.

Based on the above discussion, it can be summarized that DKDP bulk damage initiation and growth characteristics are different from the surface damage. The bulk damage initiation threshold is higher than the surface, and both the size and the number of bulk damage sites are stable under multiple UV laser irradiations with the fluence below 7 J/cm^2. In contrast, the size of surface damage does not grow, while the number of surface damage sites increases linearly with laser shots. The phenomenon suggests different mechanisms and different formations of precursors for bulk and surface damage. Clusters of holes trapped near oxygen sites are the constituent defects of the bulk damage precursors, while the source of surface damage precursors is still unknown, and there are several hypotheses including near-surface bulk defects, imbedded particles from the finishing process, and surface contamination. Microscopic inspection of the DKDP surface damage was carried out after the experiment, and two types of morphological damage sites were identified, as shown in Fig. 9.

The larger type of damage site in Fig. 9(a) consists of a rough core, cracks at the bottom of the crater, and an annulus of fractured material which is indicative of mechanical damage accompanied by spallation of material. This typical morphology is associated with a plasma fireball explosion which leads to catastrophic damage many micrometers deep with fractures. The smaller damage site shown in Fig. 9(b), which is the primary type of surface damage, is a shallow pit without observable fracture. This morphology is considered as laser cleaning more than laser damage. We suppose that this type of surface damage is caused by the absorbing precursors, which is not bound strongly enough to the crystal surface to reach high temperature and initiate a plasma fireball explosion. Thus we propose that the most likely cause of the surface damage is near-surface particles. Further experiments are planned to identify and eliminate DKDP surface damage precursors.

In conclusion, DKDP damage performance is investigated on a large-aperture laser system and is markedly different from that at small-aperture lasers. Damage occurs in the bulk and on the rear surface of the crystal under the 3ω fluences of 6.7 J/cm^2 and 3 J/cm^2, respectively. Surface damage is the major concern, not only due to the fact that its threshold is roughly a factor of two lower than the bulk, but also the number of damage sites grows linearly with laser shots. Fortunately, the sizes of surface and bulk damage keep unchanged under multiple irradiations with the 3ω fluence of 6 J/cm^2. Different damage thresholds and growth behaviors suggest different damage mechanisms and different formations of bulk and surface damage precursors. The cause of surface damage is supposed to be near-surface absorbing particles.

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