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Interactions between X-ray induced transient defects and pre-existing damage precursors in DKDP crystals

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ABSTRACT

Large-aperture laser systems, currently designed to achieve high energy densities at the target location (exceeding $\sim 10^{11}$ J/m^3), will enable studies of the physics of matter and radiation under extreme conditions. As a result, their optical components, such as the frequency conversion crystals (KDP/DKDP), may be exposed to X-rays and other ionizing radiation. This in turn may lead to a change in the damage performance of these materials as they may be affected by radiation-induced effects by either forming new damage initiation centers or interacting with the pre-existing damage initiating defects (so-called damage precursors). We present an experimental study on the laser-induced bulk damage performance at 355-nm of DKDP crystals following X-ray irradiation at room temperature. Results indicate that the damage performance of the material is affected by exposure to X-rays. We attribute this behavior to a change in the physical properties of the precursors which, in turn, affect their individual damage threshold.

Keywords: point defects, radiation-induced effects, X-rays, optical materials, laser damage

1. INTRODUCTION

Potassium dihydrogen phosphate, KH_2PO_4 (KDP) and its deuterated analog, DKDP, are technologically important and unique materials for use in high-power, large-aperture laser systems for various applications (e.g., electro-optics switching, polarization smoothing and nonlinear optical frequency conversion).[1–4] The use of nonlinear optical materials in such laser systems is continuously expanding and the laser output and frequency conversion efficiency rely on increased laser intensities. As a result, laser-induced damage in these materials represents a key limiting factor. Localized damage initiation in KDP/DKDP crystals is currently attributed to either impurity nanoparticles incorporated during crystal growth or clusters of intrinsic defects that form during growth.[5–7]

Many impurities and intrinsic stoichiometric defects have been identified in KDP/DKDP with chemical analysis, spectroscopic methods, and modeling, but efforts to correlate poor damage performance to their presence in increased relative concentrations have been inconclusive.[8–12] On the other hand, individual point defects cannot absorb sufficient amount of energy to initiate localized breakdown in these materials.[6] Therefore, clusters (aggregations) of intrinsic point defects, on the order of 10 to 100 nm, are considered to be the damage precursors (i.e., absorbing structures) responsible for damage initiation in KDP/DKDP crystals.

Previous experimental studies have shown that exposure of KDP/DKDP crystals to X-rays leads to generation of various point defects. The identity and thermal decay pathways of a number of these defects have been established by either optical techniques or electron paramagnetic resonance (EPR) spectroscopy.[12–20] Hydrogen atoms, oxygen vacancies, self-trapped holes, and holes trapped adjacent to hydrogen vacancies are among these defects. These earlier results confirmed that most of the radiation-induced point defects in the pure material are short lived above 200 K and revealed the complex relaxation pathways as one defect species decays and the electrons or holes are trapped at a different lattice site to form a new defect species.[12] However, these investigations have not established a correlation between the presence of X-ray induced defects and the laser induced damage performance of KDP/DKDP materials, which is the motivation of the present study.

In this work, we investigate the bulk damage performance at 355-nm of X-ray irradiated (at room temperature) DKDP crystals as a function of exposure time and X-ray photon energy. The goal is to understand how X-ray

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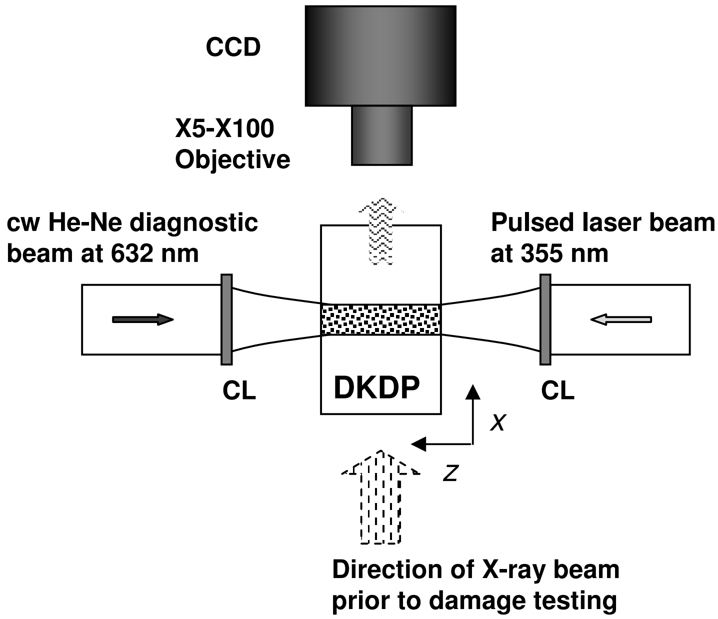

Figure 1. Schematic view of laser damage testing in the bulk of DKDP samples, both prior to and after X-ray irradiation.

induced transient point defects can affect the laser damage behavior in DKDP material by either forming new damage initiation centers or interacting with the pre-existing damage precursors.

2. EXPERIMENT

A schematic view of the laser-induced damage testing system is presented in Fig. 1 and has been described in details elsewhere.[21] The third harmonic (at 355-nm) of a $\sim$3 ns pulsed Nd:YAG laser is focused by a 200-mm focal-length cylindrical lens (CL) into the bulk of the crystal samples. We thus obtain a slit beam with a near-Gaussian profile at focus and dimensions of $\sim$50 μm in width and $\sim$3 mm in height. Scattered light images of each tested volume (obtained using a counter-propagating cw He-Ne laser diagnostic beam) are captured orthogonally to the laser propagation direction z, and the number of damage events per unit volume, or damage pinpoint density (PPD, in mm^{-3}), is measured over the region of the crystal exposed to only peak fluence ($\sim$0.12 mm^3). In Fig. 1, for illustration purposes, the bottom arrow (dashed background) and top arrow (wavy background) indicate the front surface of the sample with respect to the X-ray source (the penetration depth is measured along x) and the He-Ne light scattering by damage pinpoints induced in the bulk of the material, respectively. The damage performance of the material is quantified using two methods. First, we experimentally obtain the damage density profiles (PPD versus damage testing fluence at 355-nm) which provide a more detailed description of the damage performance over a wide range of laser fluences. The second method involves measuring the PPD at a fixed damage testing fluence as a function of location and/or post-processing parameters.

Two different X-ray tubes (with Cu and Rh targets, operating at 40 keV, 30 mA and 100 keV, 3mA, respectively) were chosen to irradiate several DKDP samples at ambient conditions. The typical energy distribution of continuous X-ray emission spectra[22] is depicted in Fig. 2 for both sources along with the estimated absorption depth of X-rays in DKDP versus photon energy (from NIST Physical Reference Data, assuming an exponential attenuation law and a mass density of 2.332 g/cm^3). These plots suggests that $\sim$50 keV or higher energy photons from an X-ray source, such as the Rh target, will experience small attenuation in the material and thus have the potential to produce point defects nearly uniformly throughout the bulk of the samples (with thickness of $\sim$1 cm). In contrast, lower energy photons from the Cu target will mostly be absorbed within a few millimeters from the front surface and the X-ray induced effects, if any, may exhibit a stronger dependence on penetration depth. Indeed, our preliminary experiments on samples irradiated with the Cu-target X-ray tube for 16, 40, and 72 hrs exposure times have indicated a small variation in the damage performance of the irradiated DKDP material as a function of testing location along the direction of the X-ray beam, i.e., proximity to front surface versus rear

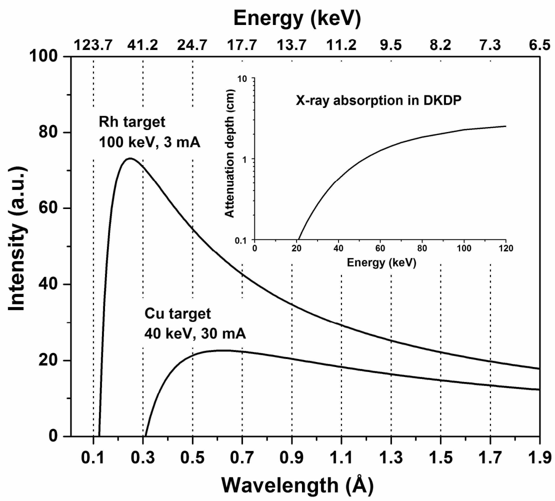

Figure 2. Typical X-ray spectra from Cu and Rh targets, $\lambda(\text{Å})=12.366/E(\text{keV})$ (from Ref. 22). (inset graph) Estimated X-ray tion depth in DKDP material versus photon energy.

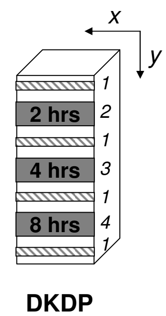

Figure 3. Damage testing regions after each X-ray irradiation step in a sequence of four: 1) 0-hr irradiation (pristine material, 4 strips), 2) 2-hr irradiation, 3) 4-hr irradiation, and 4) 8-hr irradiation (total exposure).

surface. However, these results were not consistently reproduced among a set of five samples under investigation, most probably due to sample inhomogeneities that were masking any possible X-ray induced effects, as discussed below. At this point, we decided to proceed with a careful selection of homogeneous samples based on damage testing from pristine material (prior to irradiation) as well as a more efficient irradiation scheme using the higher energy photon X-ray source (Rh target) and placing the samples ∼2 inches away for 2 hrs up to 8 hrs exposure time.

The DKDP samples investigated here were 70% deuterated, harvested from a tripler-cut plate of conventionally grown material and polished to optical quality on all sides.[4] We prepared several cubic and rectangular samples with dimensions of $\sim 1\times1\times1$ cm^3 and $\sim 1.5\times1\times5$ cm^3, respectively. Previous studies of laser-induced damage in KDP/DKDP crystals have indicated that growth conditions, raw material, and other variations can greatly affect the damage performance of these materials.[8,23] Large variations in damage densities have also been observed after testing within the same crystal boule, e.g., across growth and sector boundaries.[9] Therefore, damage testing in pristine material is a necessary step to provide the measurement baseline and factor out growth-related inhomogeneities in the damage performance of the samples. However, there is always a statistical spread in the damage density values recorded at neighboring locations, for any given fluence. Figure 3 illustrates how a rectangular sample was partitioned for damage testing to provide information prior to and after each X-ray irradiation step in a sequence of four, namely: 1) 0-hr irradiation (pristine material, tested at four locations along its height), 2) 2-hr irradiation, 3) 4-hr irradiation, and 4) 8-hr irradiation (total exposure).

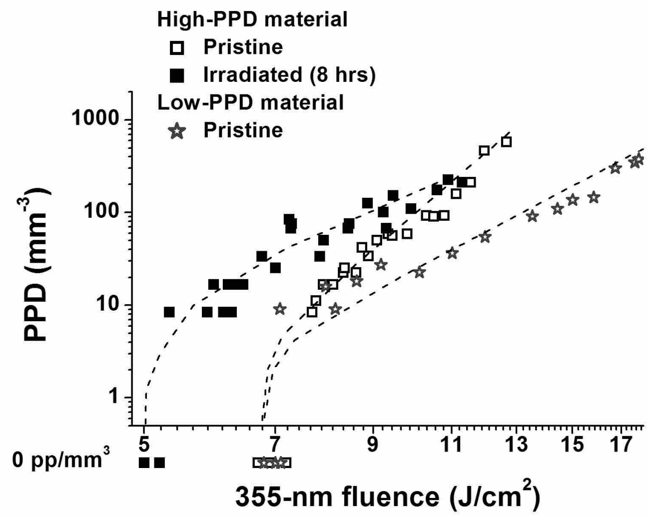

Figure 4. Damage density profiles at 355-nm measured in pristine and irradiated (for 8 hrs) high-PPD material and pristine low-PPD material. Dashed lines through the data points are drawn as a guide.

3. RESULTS AND DISCUSSION

As discussed in the previous section, we first evaluated our samples based on damage tests performed prior to irradiation at several bulk locations along x, the direction of X-ray beam propagation. We were thus able to select a set of homogenous samples exhibiting uniform damage densities (within 15%) at a fixed testing fluence. Within this batch of samples, the tests revealed two types of pristine DKDP material, with low and high PPDs at similar 355-nm fluences. As illustrated in Fig. 4, the distinct damage density profiles from each type of material exhibit a similar damage threshold at $\sim$7 J/cm^2, but the rising slopes of PPD with increasing fluence is very different.

The second step in our investigation was to determine whether or not the X-ray induced effects on the bulk damage characteristics depend on the penetration depth due to decreasing dose of radiation, i.e. compare damage test locations along x. Due to limited sample volumes, we performed this set of measurements by monitoring the PPD at a fixed laser damage testing fluence both prior to and after irradiation at a few locations throughout the sample thickness. Fig. 5 illustrates the local damage density following testing at $\sim$11.2 J/cm^2 in high-PPD material versus location in pristine and 2-hr irradiated material. These values represent the average of four measurements at neighboring locations within 1 mm, with the standard deviation indicated by the error bars. Within experimental errors, the data shows that the damage density from irradiated material is uniform throughout the thickness of the sample, suggesting the attenuation of X-ray photons is not significant over 1-cm of material. This result was also confirmed for low-PPD type samples. These results suggest that $\sim$50 keV and higher energy photons from the Rh target are responsible for the observed effects. This may also explain the low efficiency of the Cu target X-ray source in producing a measurable effect at much longer exposure times as compared to those employed with irradiation by the Rh target source.

For the remaining discussion, based on the above results, we will only report the average values of the damage densities as a function of X-ray exposure time, i.e. independent of testing locations. For the case of high-PPD material, we performed damage testing at 11.2$\pm$1 J/cm^2 in pristine (0 hrs) and irradiated material (2, 4 and 8 hrs) and the results are summarized in Fig. 6. These results demonstrate an increase in the average damage density of irradiated material vs pristine material. In addition, this effect is most pronounced for 2-hr exposure and is reduced with increasing exposure time. Furthermore, we have observed a shift of the entire damage density profile to lower fluences after exposure (the damage profile after 8-hr exposure is illustrated in Fig. 4), indicating a decrease in the overall damage performance of irradiated high-PPD material. The samples consisting of low-PPD material have been irradiated for 2 and 8 hrs, respectively. As the results in Fig. 6 suggest, for two different damage testing fluences, the effect is reversed from that observed in the high-PPD material, namely the

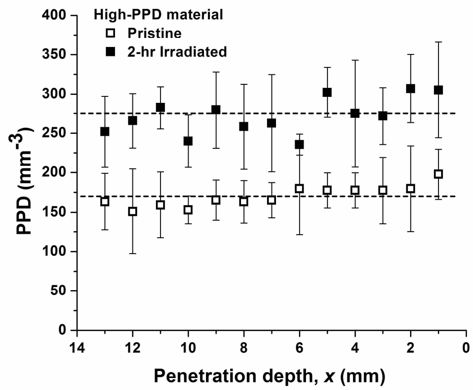

Figure 5. Damage pinpoint density versus position upon testing at 11.2 J/cm^2 in pristine and 2-hr irradiated high-PPD material. Dashed lines through the data illustrate the average values.

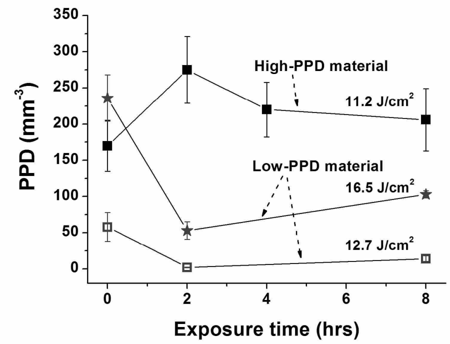

Figure 6. Average damage pinpoint density versus exposure time after testing at fixed fluences (as noted on the graph) in both pristine (0 hrs) and irradiated high-PPD and low-PPD material.

damage resistance of low-PPD material is improved upon irradiation with X-rays but the effect is again partially reversed with prolonged exposure, similar to the previous result in high-PPD material.

4. SUMMARY

The results presented here indicate that the damage performance of DKDP material is affected by exposure to X-rays but the effect is reduced with increasing exposure time. In addition, there is no evidence that new damage precursors are formed from exposure to X-rays since, for low-PPD material, an overall improvement in the damage characteristics has been observed. Instead, the results suggest that there are interaction effects between X-ray induced short-lived (transient) point defects[12–20] and the pre-existing damage precursors. This interaction is manifested as either increase or decrease in the individual threshold for damage initiation of the precursors. More recent experimental results support the model that the damage precursors are clusters of intrinsic material defects which act as multi-level absorbing particles.[7] The absorbing nanoparticle model of damage initiation predicts that the size of the precursors determines their individual damage threshold, i.e., smaller precursors initiate damage at a higher fluence.[5,6] More recently, Spaeth et al. (unpublished) have proposed that the defect density of the cluster precursors may play a key role. Therefore, the change in the individual damage threshold of the precursors can be attributed to changes in their properties that govern their ability to initiate damage, namely, a) the electronic structure of the constituent atomic defects, b) the size of the precursor and, c) the density of the constituent defects within the precursor.

The experimental results suggest that the outcome of the above interactions depends on the initial properties of the precursors, which are different for the low- and the high-PPD materials. Arguably, the electronic structure of the precursors is the same in different DKDP materials. By testing at a fixed fluence, we effectively probe the number of damage precursors which have an individual damage threshold fluence lower than or equal to that test fluence. It then follows that the lower damage performance material (the high-PPD type) contains more dense defect clusters as compared to those from low-PPD material, for a given cluster size. Upon X-ray irradiation, trapping of transient defects at the damage precursor location may lead to an increase in its size (and density, to a smaller degree) and thus lowering of the damage threshold of the precursors,[6] which is manifested as a higher PPD when testing at a fixed fluence as shown in Fig. 6. In order to explain the improvement in the damage performance of the low-PPD material following X-ray irradiation, we may need to assume that trapping of transient defects by its less dense precursor defect clusters leads to conformational changes with the formation of clusters of increased density but smaller size. These modified clusters will exhibit increased damage thresholds (due to their smaller size) compared to those prior to X-ray irradiation. Although the exact interaction mechanisms between transient X-ray induced point defects and pre-existing clusters of defects leading to damage initiation in DKDP crystals may be undetermined, this work provides a first account of such interactions not only in DKDP crystals but in any optical material.

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