Morphology of laser-induced damage of lithium niobate and KDP crystals

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The regularities of laser-induced damage are studied for $LiNbO_3$ and KDP single crystals. It is shown that the shape of damage in the dielectric crystals depends on the elastic symmetry of crystal and the propagation direction of laser beam. When the beam propagates along the optic axis of those crystals, the figures of laser damage are six-point stars for $LiNbO_3$ and four-point ones for KDP crystals. For the beam direction parallel to the *X* and *Y* axes in the KDP crystal, the damage initially has a cross-like configuration, with splitting of *Z*-oriented crack into two cracks during its further evolution.

Keywords: optical damage, anisotropy, single crystals, KDP, LiNbO₃.

1. Introduction

Studies of the bulk laser-induced damage of optical materials have usually been directed towards measuring its threshold [1-4] and analyzing mechanisms of the damage [5-7]. Only few researchers have studied bulk damage morphology, when irradiating the crystals with high-power laser pulses [8–10]. It has been noted that, in the case of light beam propagation along the optic axis, the patterns of laser-induced damage have a star-like shape, if observed along the optic axis. These stars would generally correspond to the order of the corresponding symmetry axis. When we deal with the laser beam propagation directions perpendicular to the higher-fold symmetry axis, the literature data become ambiguous. For example, YOSHIDA et al. [10] have shown that the laser beam directed along the X axis in potassium dihydrogen phosphate crystals (KH_2PO_4 or KDP) yields in eight-point damage stars. However, YOSHIMURA et al. [9] have observed six-point damage stars in $CsLiB_6O_{10}$ crystals for the identical experimental geometry, though those crystals have exactly the same symmetry as KDP (point group 42m). The goal of this work is to study regularities of the laser-induced damage in anisotropic materials by the example of dielectric crystals.

2. Experimental results and discussion

For studies of the optical damage, we used a setup described earlier in [4] and a pulsed Nd³⁺ laser (the pulse duration $\tau = 6$ ns and the output radiation energy of 60 mJ). The laser beam was focused onto the spot of about 108 µm in diameter, corresponding to the intensity level $1/e^2$. The focus plane was positioned at a depth of 3–5 mm from the sample face. For each laser beam direction, we performed several laser shots, shifting the sample after each shot in order to avoid damage accumulation. To study the bulk damage anisotropy, we used lithium niobate (LiNbO₃ or simply LN) and KDP crystals.

Laser beam intensities used for damage were fitted experimentally for both crystals, while the threshold level has not been determined because it is not the aim of the present study (readers may refer to studies [10-12]). The laser beam has passed through the crystalline sample parallel to the optic axis. We have increased the radiation energy gradually and observed whether damage occurs. After passing the damage threshold, the damage shape and dimensions have been analyzed. For small intensities exceeding $E = I/I_{\text{thr}}$ over the threshold level I_{thr} ($E \le 2$), the damage shape has been mainly irregular, with a central dark spot and randomly oriented small cracks. Regular cracks have been feebly marked. Damage dimensions have had the size of the order of 0.1 mm or less. For the excess values E > 2, the damage stars have been well marked, while the central spot dimensions have varied slightly and remained of the order of 0.1 mm. For the intensity exceeding the value $E \approx 3$ we have observed clear damage stars with the cracks of the order of 0.5-0.6 mm. Further laser energy increase leads to enlarging the damage dimensions and joining and overlapping the neighbour damaged site's cracks. Therefore, the most convenient laser beam intensity corresponds to the case of intensity excess over the threshold level $E \approx 3$. The intensity values corresponding to the specified excess over the threshold level have been used in the case of laser-induced damage presented below for the crystals under test.

The LN sample was cut out perpendicular to the principal $\{100\}$ directions. It had a shape of parallelepiped with dimensions of $8a \times 10b \times 8.3c$ mm³. The light propagated along the Z axis. The resulting damage is shown in Fig. 1. The damage stars obtained for the LN crystals, manifest nearly hexagonal shape when viewed along the optic axis (see Fig. 1a). This configuration of cracks corresponds to the point symmetry 3m of the LN. The damage channel has an arrowhead shape, with a wide part located near the light focus point and a thinning oriented into the sample (Fig. 1b).

The KDP crystal sample had nearly cubic shape, with dimensions of $11.2a \times 11.2b \times \times 10.8c \text{ mm}^3$ and the faces (100), (010) and (001). We performed damage experiments using the light directed along all of the three principal axes. When the laser beam propagates along the optic axis (the Z direction), the damage stars have the shape of precise rectangular cross, with the cross hairs directed parallel to the X and Y axes (Fig. 2a). Our results differ from those reported in [10], where the cross-type stars rotated by 45° around the Z axis have been observed, with the crack planes (110) and (110). The difference might be explained by different choices of crystallographic frame





Fig. 1. View of the damage tracks for Z-cut LN crystals (**a**) and the same damage track observed from Y-plane (**b**). The light propagates along the Z axis.



Fig. 2. View of the damage tracks for Z-cut KDP crystal (a) and the same damage track observed from (010) plane (b). The light propagates along the Z axis.

of reference. In our case, X and Y axes are oriented along the two-fold symmetry axes. At the same time, one may find in the literature that the X and Y axes in KDP are chosen as being perpendicular to the mirror planes, which are rotated just by 45° with respect to the two-fold axes. Nevertheless, the damage stars correspond to the tetragonal group $\overline{42m}$ in both cases.

YOSHIDA *et al.* [10] have observed eight-point stars from the crystallographic plane *a* for the laser beam directed along the *X* axis. The crack projections on the *a* plane observed in this experiment subtend the angles 20°, 47° and 66°. This fact seemingly agrees with neither the orientation of symmetry elements nor the experimental geometry. However, we have noticed an interesting fact. If the authors mentioned above used the alternative crystallographic reference frame and the elementary cell parameters a = b = 10.543 Å and c = 6.959 Å [13], then it would be easy to calculate that the angle between the cell diagonals is equal to 66.8°. This correlates well with the angle between the crack projections reported by YOSHIDA *et al.* [10].

When the laser beam in our experiments is directed along the X axis ($\langle 100 \rangle$ direction), we obtain the damage stars close to regular six-point ones (see Fig. 3). The orientation of the axes, as well as the reference hexagonal star, is shown in Fig. 3a. One can see that one crack plane is perpendicular to the Z axis and the two others are symmetric with respect to it (as well as the plane (010) which contains the Z axis). However, we have noticed in this case that the damage has initially a cross-type form



Fig. 3. View of the damage tracks for X-cut KDP crystal $(\mathbf{a}-\mathbf{d})$ and the same track observed from (001) plane (e). The focus plane corresponds to: exactly the damage origin (a) and 0.15 mm (b), 0.30 mm (c) and 0.45 mm (d) behind the origin. The light propagates along the X axis.



Fig. 4. View of the damage tracks for *Y*-cut KDP crystal $(\mathbf{a}-\mathbf{d})$ and the same track observed from (001) plane (e). The focus plane corresponds to: exactly the damage origin (a) and 0.15 mm (b), 0.30 mm (c) and 0.45 mm (d) behind the origin. The light propagates along the *Y* axis.

(Fig. 3a), with the cross hairs parallel to the Y and Z axes. The damage evolution deep into the sample leads to the splitting of Z-oriented crack into two cracks (see Figs. $3\mathbf{a}-3\mathbf{d}$). In this manner, the initially orthogonal-type damage transforms to a hexagonal-type one. Quite similar damage behaviour is observed for the case of laser beam directed along the Y axis (see Fig. 4). Then, the initially Z-oriented crack splits into two and forms a hexagonal star, too.

The damage channels for the KDP crystals observed along perpendiculars to the laser beam directions are shown in Figs. 2b, 3e and 4e. Unlike the LN crystals, the damage traces in the KDP crystals have almost symmetric shape along the laser beam path. Moreover, the damage channels in the KDP possess noticeable periodically situated nodes, thus indicating a self-focusing character of the damage.

In order to explain the observed shapes of damage, we have built indicative surfaces of the "expansion factor" E^{-1} (with E being Young's modulus) and its stereographic projections for the crystals under study (see Figs. 5 and 6), using the coefficients of elastic compliances taken from [14]. From the experiment, the damage star in the LN crystals, obtained for the laser light directed along the Z axis, has the points parallel



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the LN crystals: the indicative surface (a), its stereographic projection (**b**) and *X*-cut (**c**).

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to the X axis (Fig. 1a). However, the expansion factor in this direction does not acquire its extreme values. It would be suggested that the damage cracks are perpendicular to the directions of extreme values of the expansion factor (*i.e.*, Y axis in our case).

The analysis of the damage stars and the elastic symmetry for the KDP crystals allows one to make it clear which extrema of the expansion factor might be associated with the directions of cracks. As seen from Fig. 6**b**, the expansion factor has its minima in the directions of X and Y axes and the maxima in the directions $\langle 110 \rangle$ and $\langle 1\overline{10} \rangle$. The experiment (see Fig. 2**a**) gives us the orientations of the crack planes (100) and (010), when the laser beam is directed along the Z axis. This means that the cracks are perpendicular to the directions of minimal values of the expansion factor.

The conclusion could be confirmed by the following fact. As seen from the X-cut of the indicative surface of expansion factor for the LN crystals (Fig. 5c), the direction of minimal value is inclined by 18.2° to the plane XY. Thus, the cracks obtained with the laser beam directed along the Z axis must be oblique to it. It leads to wide star paths observed experimentally on the Z-cut of LN crystals (Fig. 1a). Unlike in the case of the LN, the directions of minimal values of the expansion factor in KDP are

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perpendicular to the Z axis (see Fig. 6). Consequently, the cracks really observed on the Z-cut of KDP crystals are sharp enough (see Fig. 2a).

Therefore, the results mentioned above enable us to conclude that the shape of the damage related to the laser beam directed along the optic axis in optically uniaxial crystals is explained by the elastic symmetry of those crystals and corresponds to the order of the symmetry axis. As some directions in crystals are more "compliant" and the other "stiffer", the crack orientations are not random. As a result of symmetry, such directions repeat N times per revolution, where N is the order of the symmetry axis (see Figs. 5b and 6b). Thus, the laser-induced damage cracks form symmetric stars defined eventually by the elastic symmetry of crystal.

When the laser beam propagates perpendicular to the optic axis in optically uniaxial crystal, the figures of damage cracks are more complicated. First, the damage tracks depend on the direction of laser beam ($\langle 100 \rangle$ or $\langle 110 \rangle$). Second, the symmetry of the damage pattern does not correspond to the symmetry of crystal in the given direction. Although the damage begins with appearance of two perpendicular cracks, its development leads to splitting one of the cracks (which is parallel to the *Z* axis) into two cracks and forming a hexagonal star.

3. Conclusions

Our experimental results indicate that the shape of laser-induced damage of the dielectric single crystals depends on the elastic symmetry of crystal and the propagation direction of the laser beam. When the latter propagates along the optic axis in optically uniaxial crystals, the figures of the laser damage are star-like, with the number of points defined by the highest-order symmetry axis available in the crystal structure.

References

- SWAIN J., STOKOWSKI S., MILAM D., RAINER F., Improving the bulk laser damage resistance of potassium dihydrogen phosphate crystals by pulsed laser irradiation, Applied Physics Letters 40(4), 1982, pp. 350–2.
- [2] NAKATANI H., BOSENBERG W.R., CHENG L.K., TANG C.L., Laser-induced damage in beta-barium metaborate, Applied Physics Letters 53(26), 1988, pp. 2587–9.
- [3] FURUKAWA Y., MARKGRAF S.A., SATO M., YOSHIDA H., SASAKI T., FUJITA H., YAMANAKA T., NAKAI S., Investigation of the bulk laser damage of lithium triborate, LiB₃O₅, single crystals, Applied Physics Letters 65(12), 1994, pp. 1480–2.
- [4] VLOKH R., DYACHOK YA., KRUPYCH O., BURAK YA., MARTYNYUK-LOTOTSKA I., ANDRUSHCHAK A., ADAMIV V., Study of laser-induced damage of borate crystals, Ukrainian Journal of Physical Optics 4(2), 2003, pp.101–4.
- [5] KOLDUNOV M.F., MANENKOV A.A., POKOTILO I.L., Thermoelastic and ablation mechanisms of laser damage to the surfaces of transparent solids, Quantum Electronics 28(3), 1998, pp. 269–73.
- [6] KOLDUNOV M.F., MANENKOV A.A., POKOTILO I.L., Efficiency of various mechanisms of the laser damage in transparent solids, Quantum Electronics 32(7), 2002, pp. 623–8.
- [7] EMEL'YANOV V.I., ROGACHEVA A.V., Spatial self-organisation of a defect generation wave and laserinduced formation of ordered and crystallographic-oriented regions of optical damage in crystals, Quantum Electronics 34(6), 2004, pp. 531–6.

- [8] SALO V.I., ATROSHENKO L.V., GARNOV S.V., KHODEYEVA N.V., Structure, impurity composition, and laser damage threshold of the subsurface layers in KDP and KD*P single crystals, Proceedings of the SPIE 2714, 1996, pp. 197–201.
- [9] YOSHIMURA M., KAMIMURA T., MURASE K., INOUE T., MORI Y., SASAKI T., YOSHIDA H., NAKATSUKA M., Bulk laser damage in CsLiB₆O₁₀ crystal, Proceedings of the SPIE **3244**, 1998, pp. 106–10.
- [10] YOSHIDA H., JITSUNO T., FUJITA H., NAKATSUKA M., YOSHIMURA M., SASAKI T., YOSHIDA K., Investigation of bulk laser damage in KDP crystal as a function of laser irradiation direction, polarization, and wavelength, Applied Physics B: Lasers and Optics 70(2), 2000, pp. 195–201.
- [11] WOODS B., RUNKER M., YAN M., STAGGS M., ZAITSEVA N., KOZLOWSKI M., DE YOREO J., Investigation of Damage in KDP Using Scattering Techniques, Report LLNL, UCRL-JC-125368 (1996); https://e-reports-ext.llnl.gov/pdf/230901.pdf.
- [12] ABRAHAMS S.C., MARSH P., *Defect structure dependence on composition in lithium niobate*, Acta Crystallographica Section B: Structural Science **42**(1), 1986, pp. 61–8.
- [13] BACON G.E., PEASE R.C., A neutron diffraction study of potassium dihydrogen phosphate by Fourier synthesis, Proceedings of the Royal Society of London. Series A, Mathematical and Physical Sciences 220(1142), 1953, pp. 397–421.
- [14] SHASKOLSKAYA M.P., Acoustic Crystals, Nauka, Moscow 1982 (in Russian).

Received October 28, 2007 in revised form January 15, 2008