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Sources of strain in rapidly grown crystals of KH_2PO_4

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Due to their interesting electrical and optical properties, structural phase transitions, and ease of crystallization, KH_2PO_4 (KDP) and its isomorphs have been the subject of a wide variety of investigations for over 40 years^{1,2}. Today, KDP and its deuterated analog, KD_2PO_4 (DKDP), are widely used to control the parameters of laser light such as pulse length, polarization and frequency through the first and second order electro-optic effects^{3,4}.

Efficient operation of electro-optic devices such as Pockels cells and frequency converters requires crystals with a high degree of perfection. In particular, internal strains in the crystals generate spatial variations in the refractive index tensor through the stress-optic effect.⁵ While these phenomena are a minor issue for the small crystals typically used in laboratory research applications, the effect of strain is the limiting factor on performance in applications requiring large aperture crystals such as inertial confinement fusion⁶ and high average power laser systems⁷. De Yoreo et al.⁸ analyzed the effect of internal stresses on the refractive index tensor and quantitatively related the magnitude of the stresses to experimentally determined variations in the transmitted wave front and beam polarization in KDP and DKDP crystals grown by conventional techniques. Recently, Zaitseva et al.⁹ described a method for growing bulk single-crystals of KDP from solutions at high supersaturation which produces growth rates of ten to fifty times those obtained with conventional methods. The purpose of this paper is to describe the results of X-ray topographic studies on KDP crystals grown by this technique. We show that strain in these crystals is caused primarily by three sources: dislocations, variations in composition between adjacent growth sectors of the crystal and variations in composition between adjacent sectors of vicinal growth hillocks within a single growth sector of the crystal. We find that the compositional variations cause variations in the refractive index and induce distortion of the transmitted wave front while large groups of dislocations are responsible for strain induced birefringence which leads to beam depolarization.

Single crystal boules of KDP grown at high

Figure 1: (a) Illustration of the growth habit of a single crystal boule of KDP showing the location of the seed, dislocations and the geometry of vicinal growth hillocks. (b) Location of sector boundaries in a plate of KDP cut perpendicular to the $\{001\}$ axis.

supersaturation advance on both the $\{101\}$ (pyramidal) and $\{100\}$ (prismatic) facets of the crystal leading to a pyramidal crystal habit as shown in Figure 1a. Advance of the crystal face on both sets of facets occurs on steps generated at vicinal growth hillocks formed by dislocations emanating either from the seed (see Figure 1a) or from foreign inclusions incorporated during growth. As Figure 1b shows, the vicinal hillocks on the $\{101\}$ face have an asymmetric triangular pyramidal geometry. The sectors of the vicinal hillock with the shallowest (sector 3) and steepest (sector 1) slopes generate steps oriented along the pyramid-pyramid and pyramid-prism boundaries respectively. The crystals used for this study were 1cm thick plates oriented with the normal to the plate along the $\langle 001 \rangle$ axis and were cut from the full cross section of single crystal boules. In general, such crystals contain eight

different growth sectors corresponding to the eight $\{101\}$ and $\{100\}$ directions. Within the individual $\{101\}$ sectors, lie sub-boundaries corresponding to the division between the three sectors of the vicinal hillocks (vicinal sector boundaries) as well as the boundaries between adjacent vicinal hillocks (intervincinal boundaries).

Figure 2a shows a composite of white beam X-ray topographs of a 8.8×7.6 cm KDP crystal cut from the central portion of a boule grown at 5 mm/day along the $\langle 001 \rangle$ axis. The $\{101\}$ - $\{101\}$ boundaries are faintly visible and there is pronounced contrast between the $\{101\}$ and the $\{100\}$ sectors. This contrast is also seen in the transmitted wave front profile in Figure 2b, showing that this contrast is correlated with a variation in the optic index of refraction of the crystal, a reflection of its composition. The results support those of

Figure 2: (a) Composite white beam X-ray topograph and (b) transmitted wave front profile of an (001) plate of KDP with dimensions $8.8 \times 7.6 \times 1.0$ cm³. S- crystal sector boundaries.

previous investigators¹⁰ which showed that impurities are preferentially incorporated on the $\{100\}$ faces of KDP.

Figure 3a shows a composite of X-ray topographs of a portion of a 11.5×10.0 cm KDP crystal cut from the upper portion of a boule grown at 13 mm/day along $\langle 001 \rangle$. There are three main features to this topograph: pyramid-pyramid sector boundaries (S), numerous groups of dislocations (D) and a set of domain-like structures with rectilinear boundaries which correspond to vicinal sector boundaries (V) and intervincinal boundaries (I). The shallowest sector exhibits the highest contrast relative to the other two sectors indicating that its

Figure 3: (a) Composite white beam X-ray topograph and (b) depolarization profile of an (001) plate of KDP with dimensions $11.5 \times 10.0 \times 1.0$ cm³. The dashed line in (b) gives the location of the topograph. S- crystal sector boundaries, V- vicinal sector boundaries, I- intervincinal boundaries, D- strong dislocation bunches, and T- tops of growth hillocks.

lattice parameters are the most dissimilar. Smol'skii¹¹ suggested that the contrast was due to variations in impurity content caused by differences in the segregation coefficient for the three step directions. The effect of these defects on the birefringence of the crystal is seen in the depolarization profile given in Figure 3b. While the sector boundaries are clearly visible in this profile, only the groups of dislocations with the strongest contrast cause significant levels of beam depolarization.

The effect of vicinal sectorality on the optic index of refraction is illustrated in Figure 4 which shows a composite of white beam X-ray topographs of a crystal cut from the upper portion of an

(a)

(b)

Figure 4: (a) Composite white beam X-ray topograph and (b) static fringe interferogram of an (001) KDP plate with dimensions $8.5 \times 7.5 \times 1 \text{ cm}^3$. In the absence of bulk index inhomogeneities, the fringes would be straight and parallel. Symbols are as in Figure 3.

$8.5 \times 7.5 \text{ cm}$ boule grown at 30mm/day and the corresponding static fringe interferogram. As Figure 4a shows, vicinal sectorality is strongly pronounced in this sample. Comparison of Figure 4a and b shows that the breaks and distortions in the fringes coincide with the locations of vicinal and intervicinal sector boundaries. These results demonstrate that vicinal sectorality is strongly correlated with variations in optic index of refraction and supports the hypothesis that the contrast in the topographs is caused by differences in impurity content between adjacent vicinal sectors.

The results presented here show that optical distortion from KDP crystals can be related to defects visible with X-ray topography. Strong bundles of dislocations cause high levels of strain induced birefringence while differences in composition between adjacent sectors of the crystal as well as vicinal hillocks on the faces of the growing crystal generate variations in the optic index of refraction.

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1. For a review on crystal growth see: Rashkovich, L.N., *KDP-Family Single Crystals* (Adam Higer, New York, 1991).
2. For a review on KDP structure, properties and applications see: *Ferroelectrics*, **71** and **72**, (1987).
3. Zernicke, F. and Midwinter, J.E., *Applied Non-linear Optics* (John Wiley & Sons, New York, 1973).
4. Eimerl, D., *Ferroelectrics*, **72**, 95 (1987).
5. Nye, J. F., *Physical properties of crystals*, ch. 13 (Oxford University Press, New York, 1985).
6. J.J. De Yoreo, and B.W. Woods, in: *Inorganic crystals for optics, electro-optics and frequency conversion*, Proc. Int. Soc. Optic. Engin., SPIE, **1561**, P.F. Bordui, Ed. (SPIE, Bellingham, WA, 1991, p.50).
7. Eimerl, D., *IEEE Journal of Quantum Electronics*, **23**, 2238 (1987).
8. De Yoreo, J.J. and Woods, B.W., *J. App. Phys.* **73**, 7780 (1993).
9. N.P. Zaitseva, I.L. Smol'skii and L.N. Rashkovich, *Kristallografiya* **36**, 113 (1991).
10. Belouet, C., Dunia, E. and Petroff, J.F., *J. Cryst. Growth*, **29**, 109 (1975).
11. I.L. Smol'skii, A.A. Chernov, Yu. G. Kuznetsov, V.F. Parvov, and V.N. Pozhanskii, *Sov. Phys. Crystallogr.* **30**, 971 (1985).