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Investigations of structural defects, crystalline perfection, metallic impurity concentration and optical quality of flat-top KDP crystal

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KDP crystal grown using flat-top technique has been characterized using X-ray and optical techniques with the aim of correlating the defects structure and impurity concentration in the crystal with its optical properties. Crystallographic defects were investigated using X-ray topography revealing linear and arc like chains of dislocations and to conclude that defects do not originate from the flat-top part of the crystal. Etching was performed to quantify dislocation defects density. The crystalline perfection of the crystal was found to be high as the FWHM of the rocking curves measured at several locations was consistently low 6–9 arc s. The concentration of Fe metallic impurity quantified using X-ray fluorescence technique was approximately 5 times lower in the flat-top part which falls in pyramidal growth sector as compared to the region near to the seed which lies in prismatic sector. The spectrophotometric characterization for plates cut normal to different crystallographic directions in the flat-top potassium dihydrogen phosphate (FT-KDP) crystal was performed to understand the influence of metallic impurity distribution and growth sectors on the optical transmittance. The transmittance of the FT-KDP crystal at 1064 nm and its higher harmonics (2nd, 3rd, 4th and 5th) was determined from the measured spectra and the lower transmission in the UV region was attributed to increased absorption by Fe metallic impurity at these wavelengths. The results are in agreement with the results obtained using X-ray fluorescence and X-ray topography. Birefringence and Mach–Zehnder interferometry show that except for the region near to the seed crystal the optical homogeneity of the entire crystal was good. The laser-induced damage threshold (LDT) values are in the range 2.4–3.9 GW/cm^2. The LDT of the plate taken from the flat-top region is higher than that from the bottom of the crystal, indicating that the flat-top technique has good optical quality and is comparable to those reported using rapid growth technique. The results indicate that the structural defects, crystalline quality and impurity concentration have a correlation with the optical properties of the FT-KDP crystal.

1. Introduction

Habit modification of technologically important crystals to increase their device potential is an important area of research. Several research groups have reported methods to increase the usable volume of potassium dihydrogen phosphate (KDP) crystal, which is an important NLO crystal used in high power laser applications. For example, rapid growth technique by Zaitseva and Carman [1], monosectorial KDP growth by Bespalov et al. [2], plate shaped KDP crystals by Tatartchenko and Beriot [3], oriented KDP growth by Youping et al. [4] and Salo et al. [5], splicing technique by Shui et al. [6], and unidirectional growth method by Ramasamy et al. [7]. In this direction, we had reported growth of KDP crystal directly in phase-matching direction [8], and more recently reported a new method named "flat-top growth technique" to increase the device purpose yield of this crystal [9].

Crystal growth process is a non-equilibrium process and therefore prone to defects incorporation during growth. These can be classified as point defects such as voids, interstitials and substitutions; linear defects such as dislocations; planar defects such as stacking faults, striations, impurity banding and growth sector boundaries; and volume defects such as inclusions, bubbles and precipitates. All these defects influence the quality of the crystal,

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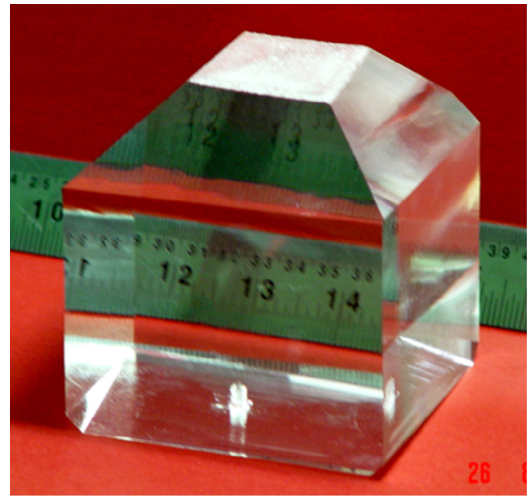
and therefore assessing their type, spatial distribution, and the history of generation and propagation during growth provide detailed information about the quality of the grown crystal, and is an essential post growth exercise. A few groups have investigated the influence of defects, impurity concentration, etc. on the optical properties of KDP crystals [10–12], however a clear understanding is still lacking. Our earlier work was aimed at reporting the flat-top growth methodology and preliminary investigations to address the question whether the flat-top growth technique introduces any defects close to the top part of the crystal. The present work is a detailed follow up of that work aimed at detailed investigations on the structural defects, crystalline perfection, metallic impurity concentration and the optical properties of FT-KDP crystal. Additionally, we have tried to establish a correlation between the defects state of the crystal and its optical properties. In order to address these issues, we have used several X-ray and optical techniques. The X-ray techniques used are the X-ray diffraction topography for imaging defects structure [13]; etching for estimating the dislocation density; high resolution X-ray diffraction for quantifying the crystalline perfection; and the X-ray fluorescence for estimating the metallic impurities [14] distribution in the crystal. The techniques used for assessing the optical quality of the crystal are spectrophotometric analysis to get an estimate of the transparency of the bulk of the crystal; optical interferometry such as the Mach–Zehnder (M–Z) and the birefringence interferometer for assessing the optical homogeneity of the sample [15]; and the laser damage threshold measurements for assessing the quality of the crystal element for high power laser applications. Section 2 describes the growth principle in brief. Section 3 describes the results obtained using various X-ray and optical techniques, and the Section 4 describes the conclusions of the present studies.

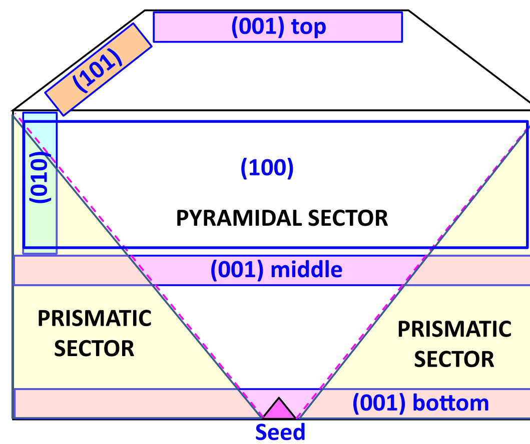
2. Solution growth of flat-top KDP crystals

KDP crystals were grown using the flat-top growth technique. The principle of the technique is as follows: A small point seed crystal was placed on a platform with the *c*-axis vertical. Growth was performed by slow cooling of the supersaturated solution. The cooling rate was successively increased from 0.02 to 0.08 °C/h depending upon the crystal surface area available for crystallization. Continuous growth for several weeks results in pyramidal portion of the crystal touching the solution–air interface. In conventional platform growth, the growth run is stopped at this stage. However in flat-top growth technique, the crystal is allowed to grow even after this stage. This results in increase of the cross-section of the prismatic sector {100} and decrease of the pyramidal sector {101}, leading to flat-top shaped KDP crystal. One such crystal of dimension $9 \times 8 \times 9$ cm^3 is shown in Fig. 1(a). The flat-top shape has the advantage that it provides enhanced volume for fabrication of device elements for electro-optic switching and frequency conversion. The crystal growth details and the empirical relationship relating the process parameters with the crystallizer design parameters have been reported elsewhere [9].

3. X-ray and optical investigations

In order to have overall picture of the defects structure of the crystal, six plates were taken from the FT-KDP crystal. The plates were normal to [100], [010] [101] and [001] directions. The location and orientation of the plates with respect to the morphology of the FT-KDP crystal (Fig. 1(b)) were such that the defects distribution of major habit faces of the crystal was covered. The results of X-ray and optical characterizations performed to investigate the defects structure, crystalline quality, metallic impurity distribution and optical quality of the flat-top KDP crystal are reported below.


(a)



(b)

Fig. 1. A flat-top KDP crystal (a) and schematic showing the location of (100), (010), (101), (001)-bottom, (001)-middle and (001)-top plates in the FT-KDP crystal obtained for X-ray and optical characterization studies (b).

3.1. X-ray topography

X-ray diffraction topography is a powerful tool for imaging defects in crystals [13]. Lattice planes around a structural defect are misoriented resulting in reduced intensity of the Bragg diffracted X-rays from such regions as compared to the regions without defect. As a result an image with varying contrast is recorded by the detector representing the defect structure of the crystal. In the present investigations a digital X-ray topography system was used which consist of a micro-focus X-ray source with Cu anode, a goniometer, a sample stage, and an X-ray CCD detector. The raw scan data was a set of linear images collected by the CCD camera as the sample was scanned across the X-ray beam. These frames were stitched by a stitching software to make stripes, which were then assembled by a tiling software to make complete topograph.

In order that the topographic image is a representative of the defect structure of the crystal under study, the sample cutting and polishing has been done very carefully to avoid introducing defects which will result in spurious artifacts in the topographs. A good sample for topography has to be optically flat and damage free surface resulting in features that are attributed to only the

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