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UCRL-JC-132196
PREPRINT

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This paper was prepared for submittal to the
Third Annual International Conference on
Solid State Lasers for Application (SSLA)
to Inertial Confinement Fusion (ICF)
Monterey, California
June 7-12, 1998

May 1, 1998

Lawrence Livermore National Laboratory

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Growth of large KDP crystals in the form of plates.

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ABSTRACT

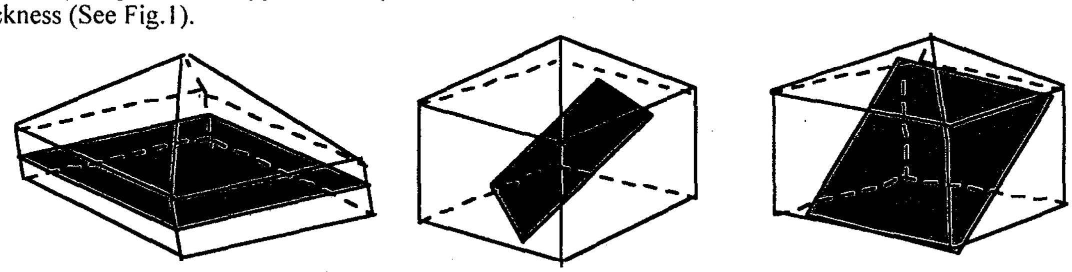
This paper suggests a new technique of growth-oriented KDP crystals in the form of plates. The technique includes: using small oriented seeds spaced between two parallel platforms with a rapid growth of crystals between these two platforms, in a tank containing a KDP solution. As a result, crystals in the form of plates can be obtained. The thickness of the crystal plate depends on the distance between platforms. The horizontal dimensions of the plate depend on the volume of solution and the diameter of the platforms. The orientation of the plates are defined by the orientation of the seed. KDP crystals in the form of plates of two orientations are grown. The peculiarities of morphology and some characteristics of crystals are discussed.

Keywords : KDP, Solution growth, Oriented crystals, Rapid growth, Shaped growth, Growth between platforms.

1. INTRODUCTION

KDP and DKDP single crystals are used for effective control of laser emission properties (intensity, polarization, wavelength, and alignment) especially in cases where the size of the cross section of the light beam is tens of centimeters. For instance, for the National Ignition Facility (NIF) and Laser MegaJoule (LMJ) projects, frequency conversion plates and Pockels cells with dimensions on the order of 410 x 410 x 10 mm are needed. In this case, there is no alternative to the KDP and DKDP crystals grown from the aqueous solution.

With respect to our information maximal dimension of plates cut from crystals grown by conventional technique is 370 x 370 x 10 mm . Therefore at present there is only one technique developed at Lawrence Livermore National Laboratory (LLNL) that allows some KDP crystals to grow to sizes sufficient to cut plates with dimensions of 410 x 410 x 10 mm [1]. But we need a very large crystal (approximately 550 x 550 x 550 mm) in order to be able to cut from it even a few plates of 10 mm thickness (See Fig.1).



a b c

Fig.1. The minimum dimensions of KDP crystals that we need to cut from them:
a. Z - plates: 41 x 41 x 41 cm (< 37% of volum of crystal are used);
b. Dubler of type 1: 52 x 52 x 52 cm (< 6% of volum of crystal are used);
c. Tripler of type II: 55 x 55 x 55 cm (< 10% of volum of crystal are used).

It is necessary 1,000 litters of KDP solution with a temperature of saturation of order 75°C for growth of this crystal. At the same time only 5-10% of crystal volume can be used for plate production and as a result the price of plates will be very high. This problem is very important especially for DKDP because of the cost of heavy water and DKDP salt. For instance, now with respect to the specification that exists for LMJ we need: 264 z - plates in KDP or DKDP for Pockel's cells (410 x 400 x 10 mm), 264 KDP Doublers type I (395 x 410 x 12 mm) and 264 DKDP Triplers type II (400 x 410 x 9 mm). To realize this program over a 4-5 year period, using the rapid-growth technique developed by LLNL, we would need 8 systems for crystal growth with a solution volume of 1,000 liters for each system. We would need also between 30 to 40 tons of KDP and/or DKDP salt and between 6 to 10,000 liters heavy water.

With respect to the agreement between the Commissariat of Atomic Energy of France (CEA) and the Department of Energy of USA (DoE) we have now a collaboration with the LLLNL Crystal Growth team which allows us to grow large KDP crystals by using the rapid-growth technique developed by LLNL (see Fig. 2). But, because of the reasons discussed above, for the past three years CRISMATEC has been developing a crystal growth technique for the orientation of the KDP plates we need.

Fig.2. Large KDP crystal grown by French team using the rapid-growth technique developed at LLNL.

2. GENERAL DESCRIPTION OF GROWTH TECHNIQUE

The problem of shaped crystal growth (a growth of crystal in the form of a plate, tube or other more complicated forms that are required) and, as a result, of the maximum use of a volume of grown crystal exists for all techniques of crystallization and especially for those from melt. For instance, using the Czoghralski or Kyropulos technique we grow a cylindrical crystal and after that we need to cut the crystal and prepare plates, tubes or other more complicated forms for industrial application. As a result we lose a lot of expensive material and spend a lot of time and money for mechanical treatment of crystals. This is the reason why there have been many attempts to develop shaped crystal growth techniques from a melt. This problem is discussed in detail in[2,3].

The comparison of problems of shaped crystal growth from a melt and from an aqueous solution shows that second problem is more complicated. The fact is that when a growth from melt is carried out an interface melt - crystal, as rule, coincides with an isotherm of the crystallazation temperature. This allows us to realize many techniques of shaped crystal growth, for instance, the technique of pulling from a shaper of crystals with very complicated form [2,3].

When a growth from an aqueous solution is carried out there are two main peculiarities of this process: the first - an interface solution - crystal is a set of singular faces of crystal producing a closed form; the second is concerned with a regeneration of seed: on the boundary of seed and grown crystal there are many defects and this volume of crystal can not be used for technical application. These peculiarities prevent a large distribution of techniques of shaped crystal growth from solutions.

From our point of view there is only one serious attempt to solve this problem : The Laboratory of Crystal Growth, from the Institute of Applied Physics, of the Russian Academy of Sciences has developed, over the last 15 years, a technique of rapid growth of large KDP plates restricted by walls of Plastic form located in solution [4]. There is serious disadvantage of this technique: a complicated pump has to operate in supersaturated solution. As a result, on a level of technique that exists now supersaturated solution can serve up to a maximum of three weeks. After that, subindividual crystals appear in solution and this phenomenon limits the dimensions of plates in the direction of growth (250mm) that can be achieved by this process.

On the basis of our experience [2,3] we tried to realize as many schemes as possible of KDP crystals growth from aqueous solution. There is one scheme that is the most fruitful. The CEA has granted a patent for this technique [5]. We shall discuss this technique below.

The idea is the following : Let us assume that we need a frequency converter of a definite type. We cut the seed in the form of small plate: 10 x 10 x 4 mm. A crystallographic orientation of surface of the seed plate coincides with one of the converter.

We prepare a system for crystal growth consisting of two parallel horizontal platforms connected by a set of vertical bars located on the edges of platforms. The distance between platforms that we used was 15 - 40 mm. The upper platform is fixed on an vertical axis (see Fig.3a). Sometimes it is possible to use some platforms forming a set consisting of some cells of growth. In this case, the upper platform of one growth cell serves as a lower platform for next growth cell.

We glue the seed (or seeds) on a surface or in small hollow of lower platform and put the two platforms (or set of platforms) in the aqueous solution of KDP salt with an equilibrium temperature of 50 - 75 °C. We realize an alternative rotation of a set of platforms and a lowering of solution temperature to obtain a supersaturation of solution on the level of

3% - 6%. A rapid three-dimensional crystal growth begins(Fig.3b). When the growing crystal approaches the higher platform, vertical growth comes to a halt and two-dimensional plate growth begins (Fig.3c) and finally we obtain a full size crystal after 30 days (Fig.3d). The set of platforms allows to realize the growth of many plates in the same tank simultaneously (Fig.3e).

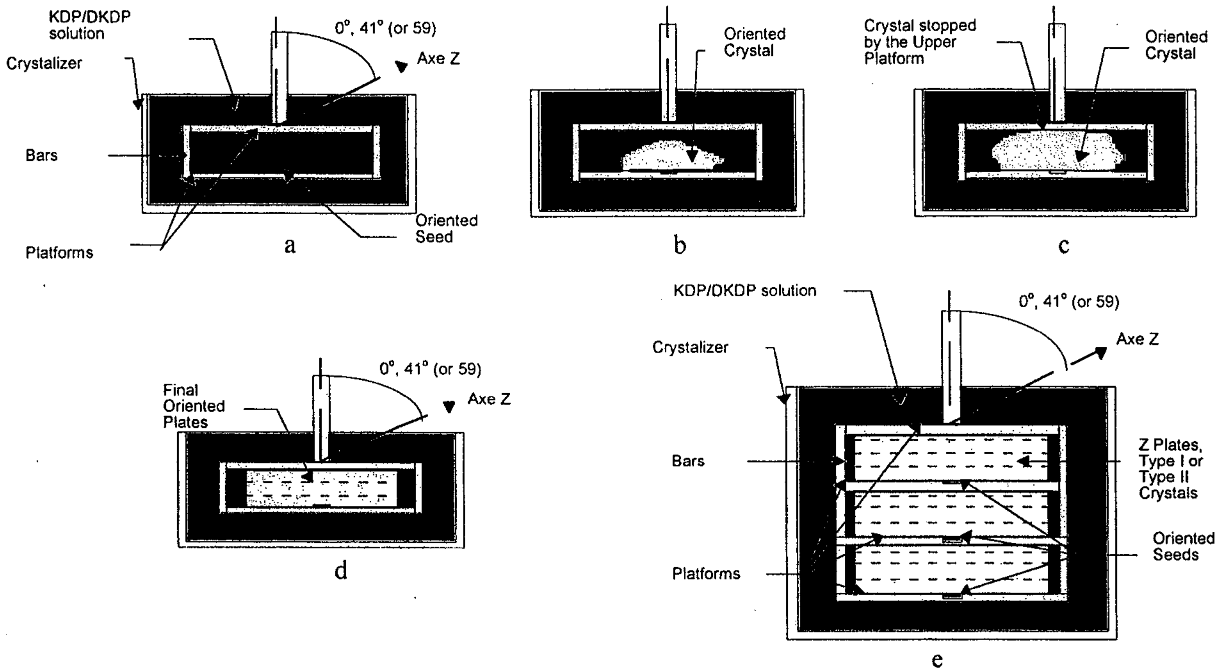


Fig.3. The technique of crystal growth between two platforms: a. A system for crystal growth consisting of two parallel horizontal platforms; b. A set consisting of some cells of growth, an upper platform of one cell of growth serves as the lower platform of next cell of growth. c. A rapid three-dimensional growth of crystal. d. The growing crystal approaches the higher platform, vertical growth comes to a halt and two dimensional growth of plate begins.
e. The set of platforms allows to realize the growth of many plates in the same tank simultaneously.

3. DISCUSSION OF RESULTS OF EXPERIMENT

As a result of our experiments, we have achieved two types of frequency converters (first and second type)for the orientation of KDP plates.

In 1996 we have grown a first plate without visible defects. This is the plate oriented as a frequency converter of type II with the dimentions: 110 x 90 x 15 mm. It is possible to see it on Fig.4a, cut for X - ray topographic investigation.

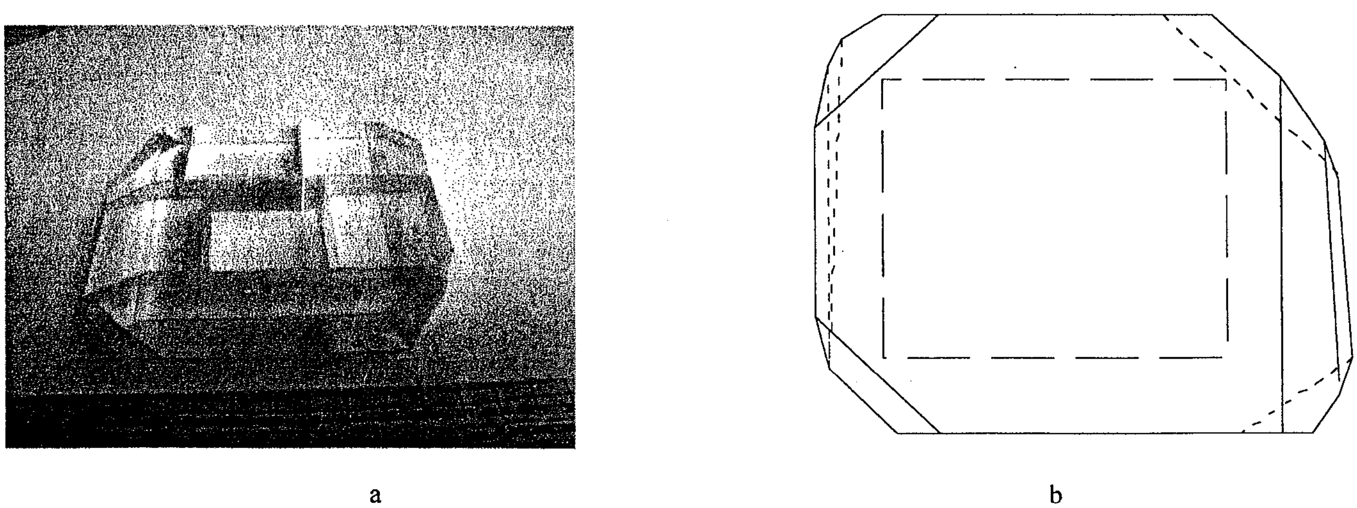
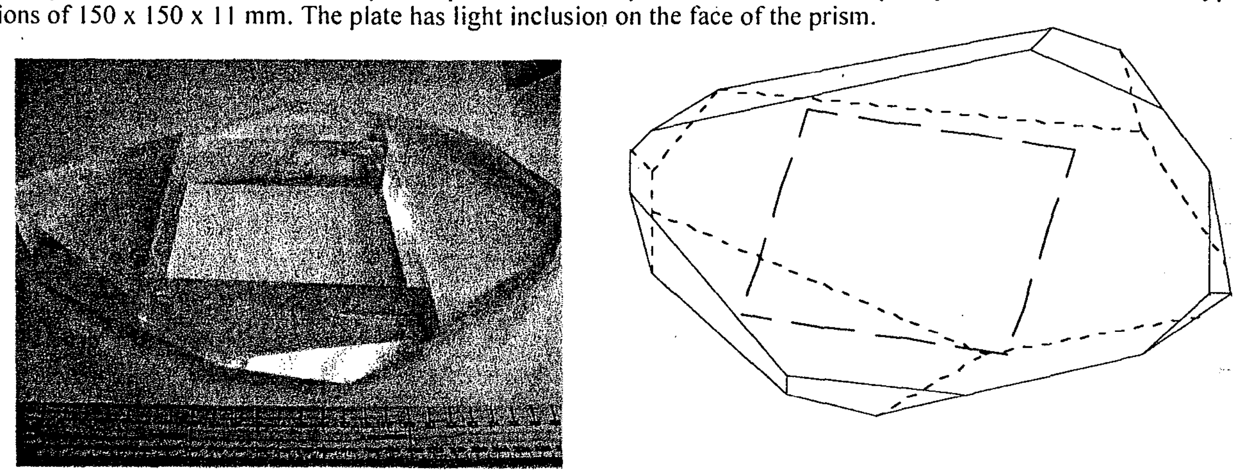


Fig.4. KDP crystal plate oriented as a frequency converter of type II with the dimention:
110 x 90 x 15 mm. a. A view of the plate. b. A configuration of the plate.

The Fig.4b shows a configuration of the plate. We marked a part of the plate that can be used for converter production. The plate has a nearly square form. All 4 prismatic faces and 6 (from a possible 8) bipyramidal faces are presented on the crystal configuration. More than 50% of the volume of crystal can be used for converter production.

During a two year period (1996 and 1997) we have grown some plates of frequency converter orientations of first and second types from the tanks of 30 liters and 60 liters. As a rule, an average speed of growth was 10 mm/day. Some of these plates do not have any visible defects.

In 1997 we have grown the biggest plate (frequency converter of the first type) that is possible to grow from a tank of 400 mm of diameter and 60 litters of volume. Fig. 5a shows the picture of plate. It has a rhomboid form 40mm thick with a larger diagonal of 360mm. The central part of plate is used for production of three frequency converters of the first type of dimensions of 150 x 150 x 11 mm. The plate has light inclusion on the face of the prism.



a b

Fig.5. The biggest KDP plate, oriented as a frequency converter of the first type, that is possible to grow from a tank 400 mm in diameter with a volume of 60 liters . a. A view of the plate. b. A configuration of the plate.

The Fig.5b shows one configuration of the plate. We have marked a part of plate that can be used for converter production (more than 30% of the volume of crystal). All 4 prismatic faces and all 8 bipyramidal type are presented on the crystal configuration but a range of faces is unusual in comparison with the configuration of crystal grown without two platforms. Here some faces of bipyramidal faces form acute angles. These angles could not be seen on a crystal grown in ordinary conditions. This interesting phenomenon has to be studied in further detail.

We began the investigation of some peculiarities of crystal plate growth between platforms. On Fig.6 the X-ray topogram of a prism face is presented. The topogram was obtained with Dr. Igor Smolsky on the Stanford University synchrotron by using white X-ray radiation monochromatized by a Mo filter. A dislocation source of growth steps exists at the central part of the sample. An inclined line presents an edge of vicinal hillock which is built by the growth steps. A natural crystallographic edge of prismatic face is clearly seen in the upper part of the topogram. Image contrast of this edge is increased as a result of the existence of stronger stresses at the sectorial boundary. The curvature of macrosteps shows a general direction of their movement from this edge to the center of the face. It allows us to speculate that this edge may be the source of the growth steps.

We were unable to realize a systematic investigation of optical characteristics of grown plates. But preliminary measurements indicate that crystal plates grown between platforms sometimes meet the LMJ homogeneity specification.

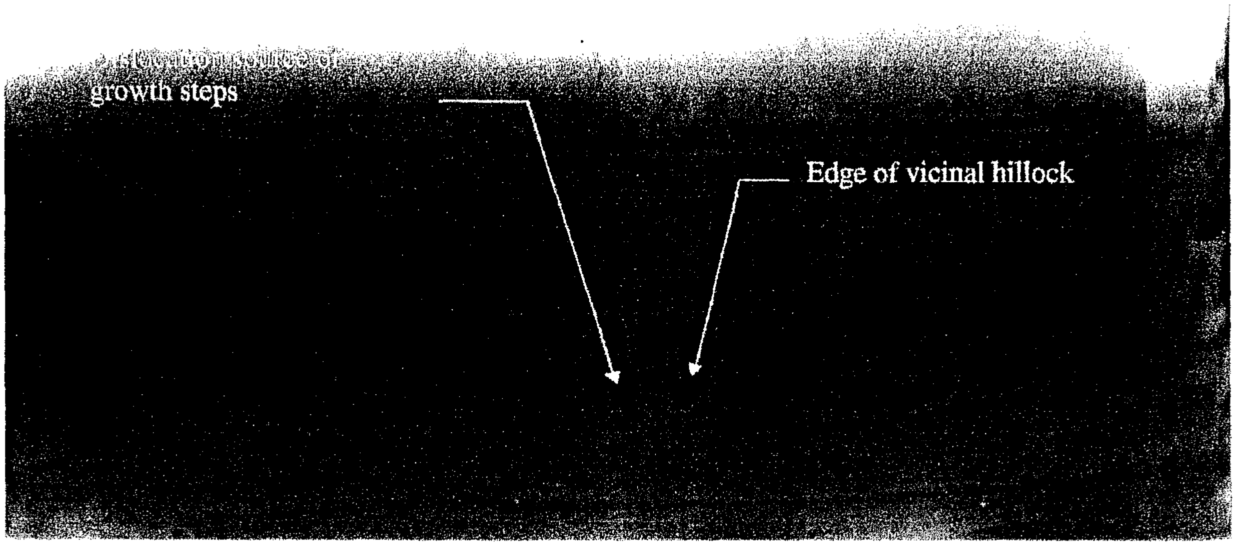


Fig.6. X-ray topogram of prism face.

4. CONCLUSION

We have proven that it is possible to grow oriented KDP crystals in the form of plates between two platforms.

Preliminary optical investigation shows that this technology is able to meet LMJ homogeneity specifications.

This technology is able to optimize the industrial and financial aspects of large crystal KDP plate production, reducing by 4 the volume of solution and by 2.5 the average cost of plates in comparison with the existing growth technique.

5. ACKNOWLEDGMENTS

The authors gratefully acknowledge the contributions of Pierrick Defossez for crystal growth assistance, Igor Smolsky for X-ray topogram assistance and Natalia Zaitseva and Igor Smolsky for fruitful discussion.

This work was performed under the auspices of the Commissariat à l'Energie Atomique (France) CEA, Departement Laser - Plasma (DLP), Service Conception Système Laser (SCSL), Laboratoire des Technologies Optique (LTO) by CRISMATEC under Contract No.3792/LV. This work was performed under the auspices of the U.S. DOE by LLNL under contract No. W-7405-Eng-48.

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