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Rapid Growth of Large-Scale (40–55 cm) KDP Crystals

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Abstract

KDP (KH_2PO_4) single crystals up to 47 cm in size have been grown by the rapid growth technique on the point seed in glass crystallizers of 1000 L in volume at growth rates of 10 to 25 mm/day in both the [001] and [100] directions. Measurements of the optical quality of 41 x 41 cm single crystal plates are presented.

1. INTRODUCTION

The work on rapid growth of KDP-type crystals, mainly KH_2PO_4 (KDP) and $K(D_xH_{1-x})_2PO_4$ (DKDP), was initiated in the early eighties by the need for large aperture single crystal plates in Nova, the world's largest laser. The frequency conversion arrays on Nova are constructed from 27 cm x 27 cm KDP crystals grown, at that time, by traditional techniques. The baseline design for Nova's successor—the National Ignition Facility (NIF) [1] incorporates about 600 41 cm x 41 cm Pockels cell, doubler, and tripler crystals. Depending on the type of frequency conversion used for the NIF, the boules which are needed to yield crystal plates of this size must be about four times the size of the Nova boules (see Fig. 1).

Conventional crystal growth usually takes place at growth rates of 1-2 mm/day for KDP crystals and not faster than 1 mm/day in the case of DKDP. At these rates, it can take more than two years to get a crystal of the size required for NIF. As Fig. 2b shows, the final size of a traditionally grown boule is approximately twice as long as it is wide because of the large portion of the crystal occupied by the regenerated seed and the material with high density of dislocations originating at the seed cap. Additionally, at the low supersaturations used in conventional growth, impurities in the starting salt generate what has been referred to as a “dead zone” [2] in which the growth rates along the [100] and [010] directions is near zero. Attempts to increase growth rates often resulted in the formation of defects on prismatic {100} faces which start growing at supersaturations just above the “dead zone”. As a result, conventional crystals are grown only in the [001] direction and the growth of the boule must be preceded by the complicated process of obtaining seeds which have the cross section of the final crystal (see Fig. 2a). This significantly increases the production time.

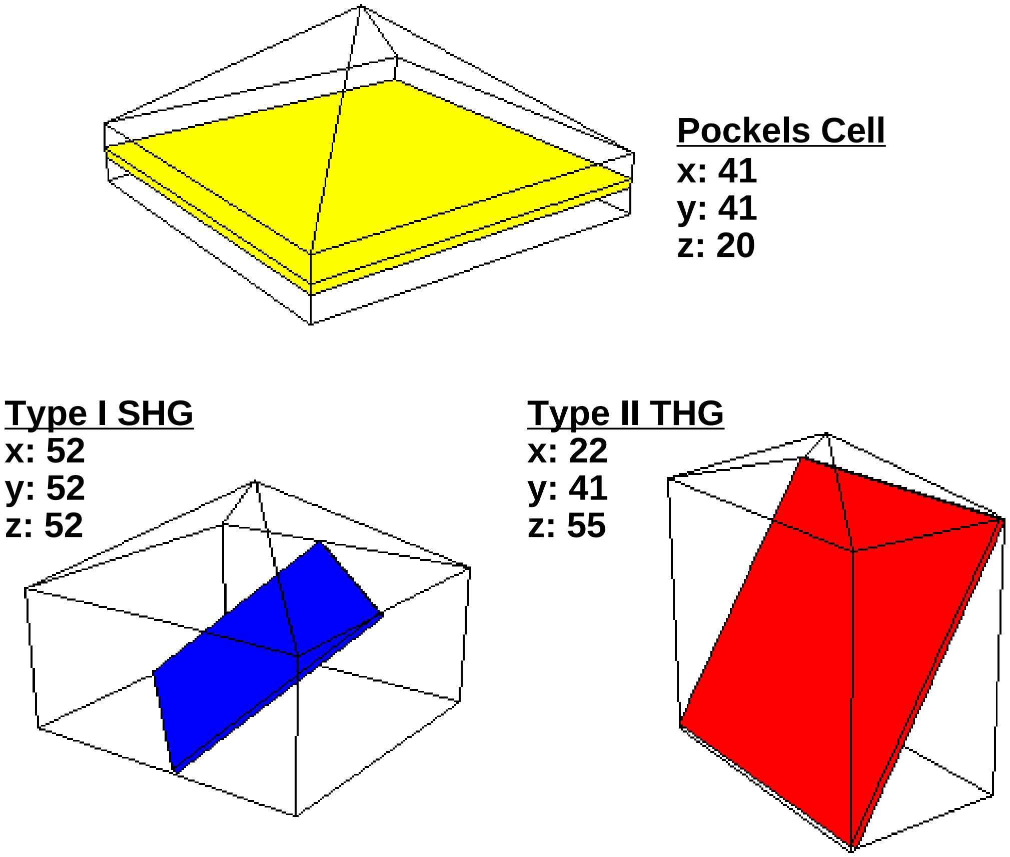

Fig. 1. The minimum size of KDP single crystal boules (in cm) needed to obtain Pockels cell and harmonic generation plates of different types for the NIF project.

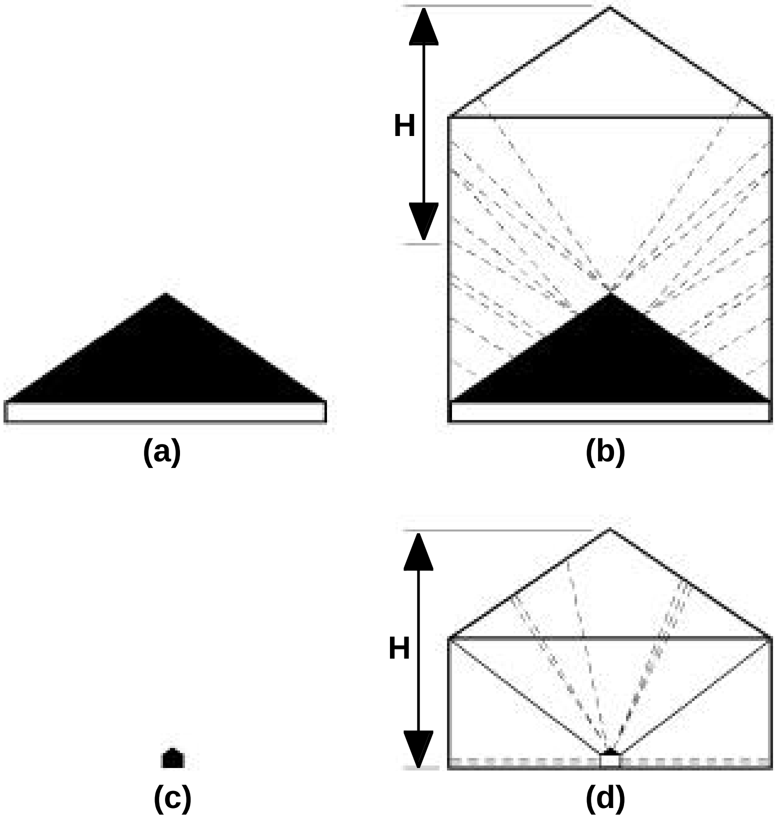

Fig. 2. Schematic of traditional crystals (a, b) and those grown rapidly on the point seed (c, d). Seeds with regenerated caps (a, c). Final crystals with the length H needed for cutting plates (b, d). Hatched regions—seeds. Solid regions—regenerated seed caps. Dashed lines—dislocations. Light solid lines—boundaries between {101} and {100} sectors.

The use of very pure salts can reduce the width of the "dead zone". As was shown previously [3], if growth is performed in a regime where the growth rates along [100] and [010] are sufficiently high and comparable to that along [001], defect formation on those faces does not take place and crystals can have high optical quality both in the prismatic and pyramidal sections.

Several rapid growth techniques [4-7] have been developed in recent years to grow KDP-type crystals at growth rates more than an order of magnitude larger than those obtained with the traditional technique. It was shown that rapidly-grown crystals can have as high optical quality as those typically grown by the traditional technique [3,8,9], but, until now, the size of rapidly-grown crystals did not exceed 15-20 cm. As a result, the 37 cm crystal plates currently in use on Beamlet, the NIF prototype, still had to be grown by the traditional method.

Here we report the growth of KDP crystals up to 47 x 46 cm^2 in cross section at rates in excess of 10 mm/day. The method used to grow these crystals is based on the use of the "point seed," described previously [10]. The small size of the seed used in this method does not depend on the final size of the crystal, because the crystal grows uniformly on both prismatic {100} and pyramidal {101} faces (see Fig. 2c and Fig. 2d). While the presence of the prismatic faces—and the resulting polysectorality—present a potential source of optical inhomogeneity in these crystals, recent measurements made on crystals grown by this technique with sizes of 10-15 cm showed that, at growth rates of 10-15 mm/day, crystals grown from high purity salts have good optical uniformity and the entire volume of the crystal can be used for obtaining high quality plates [9].

3. APPARATUS

The full-scale rapid growth system is shown schematically in Fig. 3 and in a photograph in Fig. 4. It consists of a 1000 liter glass tank placed in a controlled temperature water bath. The crystal is grown on a square, acrylic platform 60 x 60 cm^2 in size and is rotated alternately in one direction and then the other on a symmetrically programmed schedule with controlled acceleration, deceleration and rotation rates. A special device developed for this work, called a seed protector, is inserted within the platform shaft where it can freely move up and down. This device allows us to assemble the empty crystallizer with the point seed (about 1 cm^3 in size) hermetically sealed within the seed protector at room temperature. The crystallizer is heated to high temperature—about 10°C above the saturation point—the KDP solution which is mixed in a separate process tank, is then transferred into the crystallizer and solution treatment, such as filtration and overheating, is executed for any desired length of time while the seed is perfectly preserved under the seed protector. At the moment when the solution is ready for growth, the seed protector is raised up and the growth process begins. The seed protector can also be used if the growing crystal needs to be remelted. Once the crystal is melted to the size of the initial seed, it can then be covered by the seed protector and solution processing can be repeated. In this way, the growth process can be repeated several times without opening the crystallizer to introduce a new seed.

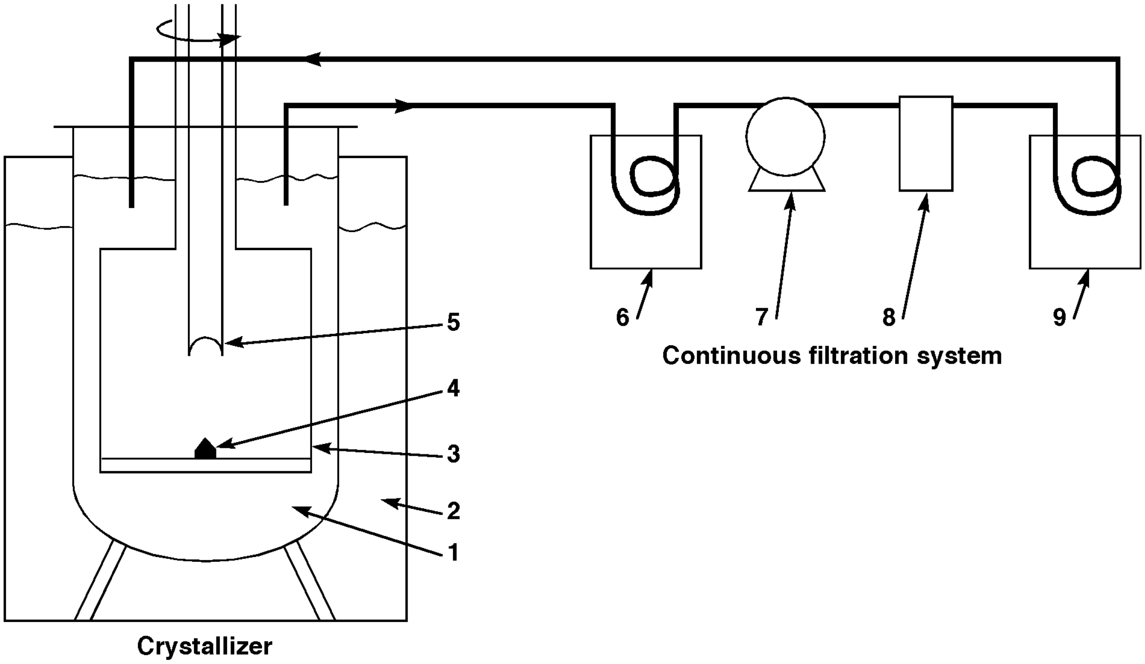

Fig. 3. Schematic of 1000 L crystallizer: (1) 1000 L glass growth tank; (2) temperature controlled water bath; (3) platform; (4) seed; (5) seed protector; (6) heater; (7) pump; (8) filter; (9) cooler.

Fig. 4. Photograph of the 1000 L crystal growth system.

Previously, Montgomery et al. [11] showed that high laser damage thresholds could be obtained in conventionally grown KDP by continuously filtering the solution during growth. The continuous filtration system shown in Fig. 3 has been developed in this work for filtration growth solutions at high supersaturation. It contains three temperature controlled sections: a superheater and filter (operating at 80°C), and the third section where the filtered solution is cooled to the growth temperature. This system has been used to grow KDP and DKDP crystals in small 20L crystallizers up to a size of 17 cm and tested in 1000L tanks where it will be used after the first 55 cm crystals have been grown.

4. SOLUTION STABILITY AND CRYSTAL GROWTH

Our previous work [12] made in small 5-20 L crystallizer showed that KDP, as well as DKDP solutions, can be stable against spontaneous nucleation for induction time periods, τ, of months at undercoolings of up to 30°C and, during continuous cooling, supersaturations σ of up to 100-130% can be reached (for practical measurements we use relative supersaturation $s = (C - C_0)/ C_0$, where C and C_0 are the actual and equilibrium concentrations expressed in weight fraction). We showed that the actual stability of growth solutions is much higher than needed for practical crystal growth (Fig. 5), as proved by the absence of spontaneous nucleation in small crystallizers (5 - 20 L in volume) during crystal growth at rates of 10-40 mm/day. The use of the seed protector device in 20 L crystallizers allows us to grow KDP crystals at the growth rates up to 60-70 mm/day. The reason for terminating growth at such high rates is not spontaneous nucleation from the solution but usually defects and finally cracks in the growing crystal which, in our opinion, are due to the effect of impurities or hydrodynamic conditions. These results show that at the supersaturations needed for practical growth rates (usually not faster than 20 mm/day) in well filtered and overheated solutions spontaneous nucleation in the solution itself is not a problem. From our

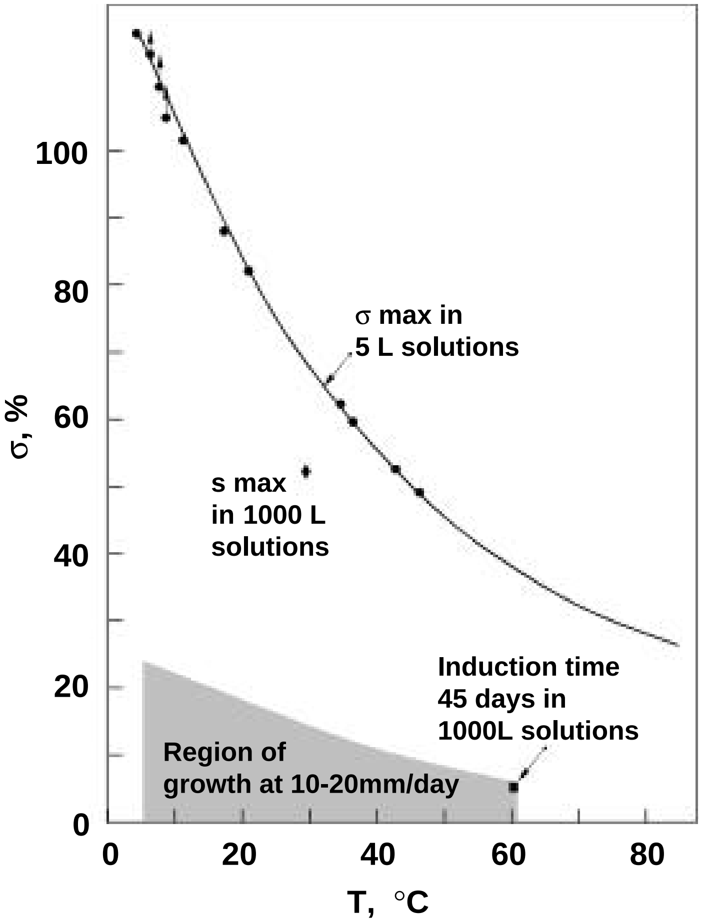

Fig. 5. Maximum stability obtained in KDP solutions in comparison to the region used for crystal growth.

observations, any extraneous crystals which do appear in the shaded region of Fig. 5 are caused by the main crystal (either through cracking or moving relative to the crystalholder) or faulty equipment, such as dry surfaces or poorly bonded joints in platforms.

Measurements made in the 1000 L tanks shown in Fig. 6 gave solution stability of the same order of magnitude: during continuous cooling, σ of about 50% was reached and the induction time τ in a stirred solution saturated at 65°C was more than 1.5 month at $\sigma = 6\%$ (the run was interrupted to continue crystal growth experiments).

The initial saturation temperature of the solutions in different growth runs varied from 60 to 68° C. The saturation temperature does not need to exceed 70° C to obtain full size crystals of about 55 cm on a side and 250 kg in weight from solution volumes of 1000 L.

Crystals were grown at rates of 10-25 mm/day along Z axis and 5-12 mm/day along the X-Y directions. Supersaturation and required cooling rate were controlled by measuring growth rates and crystal dimensions during the growth process.

More than ten KDP and one DKDP crystal with sizes of about 40-47 cm in all directions have been grown. In the case of the DKDP crystal, the initial saturation point of the solution was 66° C, the deuteration level of the solution was about 92%, the growth rate along Z axis was about 14 mm/day and each prismatic face grew at a rate of 10-12 mm/day. Precipitation of the monoclinic phase was never observed during the growth of the tetragonal crystal, which once more demonstrates high stability of the growth solutions against spontaneous nucleation.

One of these large KDP crystals grown in a 1000 L crystallizer is shown in Fig. 6 in comparison with a typical 12 cm crystal grown in a 10 L crystallizer. The weight of the large crystal is about 130 kg compare to 1.5 kg for the smaller one. Both crystals were grown on point seeds of the same size. The dimensions of the large crystal are about 45 cm in all X, Y, and Z directions. The entire period of growth was 30 days.

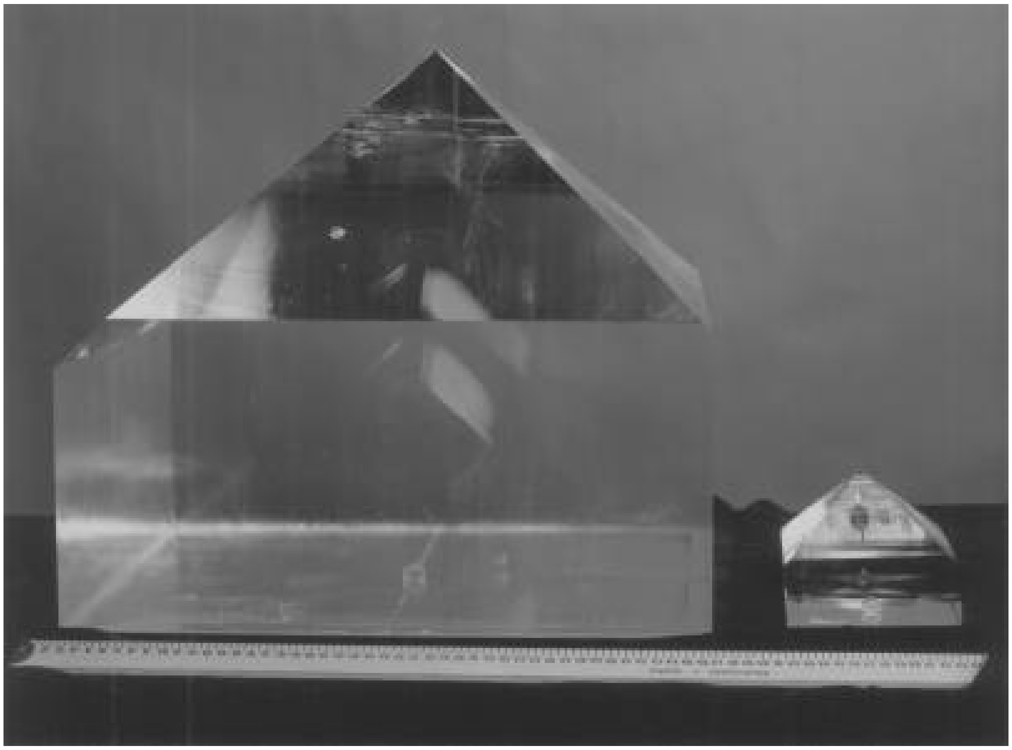

Fig. 6. Photograph of KDP crystals grown in 1000 and 10 L crystallizers.

At present, the main obstacles in obtaining crystals of the larger size are connected with mechanical problems resulting from the large size of the equipment, such as deformation of the platform-crystalholder in response to flow of the solution which is more than 1000 kg in mass, or the method of supporting the growing crystal which is initially connected to the platform only through the 1 cm^3 seed. These problems are still being solved, but even the crystals currently being grown have sufficient size to provide Z-plates of NIF size.

5. CRYSTAL QUALITY

Only preliminary measurement have been performed on the crystals grown in 1000 L crystallizers. Some crystals had no visible defects, though in many crystals solution inclusions appeared randomly on both prismatic and pyramidal faces. These defects typically were slight on the prismatic {100} faces and much more pronounced on the top of the pyramidal {101} faces. The nature of these defects is not yet clear and is a subject of current investigations.

The surface structure of the crystals did not differ from that typically observed in the small crystals: each prismatic face typically had only one growth hillock clearly seen in reflected light from the beginning to the end of growth. Growth hillocks on the pyramidal faces changed more often. One or two hillocks with intervicinal boundaries could typically be observed at any time on the same face, which is also typical for small crystals [13].

One of the large crystals which had a crossection of 44 x 44 cm^2 was cut into Z [001] plates from which two 41 x 41 x 1 cm Pockels Cell crystals were fabricated (see Fig. 1). Measurements of anomalous birefringence and transmitted wavefront distortion on these plates, as well as measurements of the laser-induced damage threshold on witness samples taken from adjacent positions in the boule show that the optical quality of these crystals meets the NIF requirements.

In Fig. 7 we show a profile of the depolarization loss measured in a 29 x 29 cm section of one plate. The average loss is 4×10^{-2}% and the maximum loss is about 0.1%. These numbers are five to ten times smaller than what is specified for the NIF. Fig. 8 shows the distortion of the transmitted wavefront due to index variations in the bulk of this plate. The peak-to-valley distortion is $< \lambda/4$ and the maximum gradient is $< \lambda/10\ cm^{-1}$. Both of these values meets the NIF specifications. The measured laser conditioned damage threshold at the first harmonic (1064nm) using 7.6ns pulses is $32 J/cm^2$. When scaled to 3ns, this value exceeds the NIF requirement for both the Pockels Cell and the 2nd harmonic generator. Our preliminary measurements also show no direct dependence of the damage threshold on sectorial structure of the crystals grown on the point seeds [14]. All this demonstrates once more that the presence of the prismatic sectors and sector boundaries are not the obstacles for obtaining large single crystal plates of high optical quality as was expected many years ago when this work just begun [15].

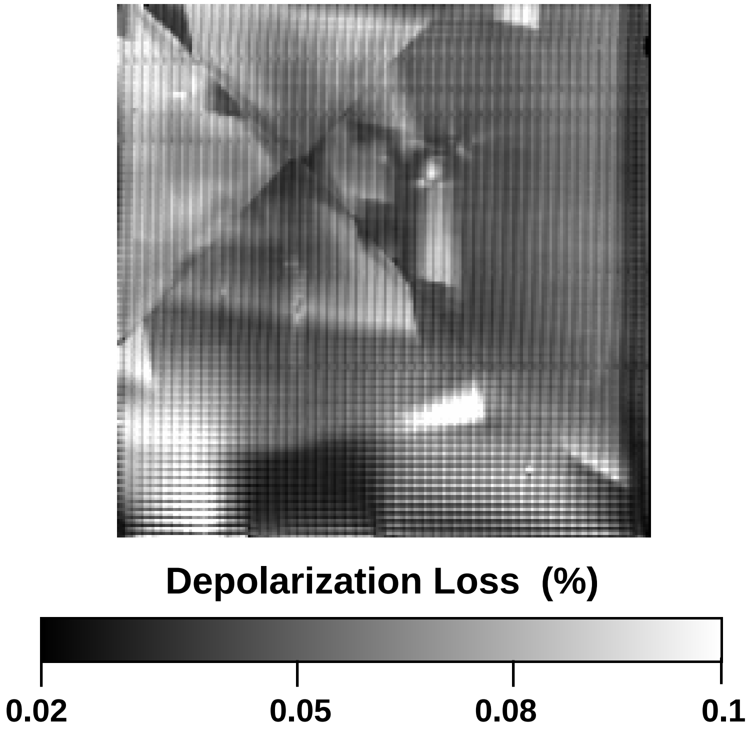

Fig. 7. Profile of the depolarization loss in a 29 x 29 cm^2 section of a 41 x 41 cm^2 plate. Sector boundaries and growth hillock boundaries are clearly visible but result in minor depolarization losses.

6. CONCLUSION

In conclusion, our results show that large KDP single crystals for laser fusion systems can be grown rapidly using high supersaturations. The next task of this work is to develop a stable technique for obtaining KDP and DKDP crystals up to 55 cm in size which have high optical quality including laser damage threshold. High reproducibility of the results on growth and optical quality which is typical for the small rapidly grown crystals and which has been demonstrated at large scale leads us to believe that the development of the rapid-growth technique which was initiated more than ten years ago is close to its conclusion.

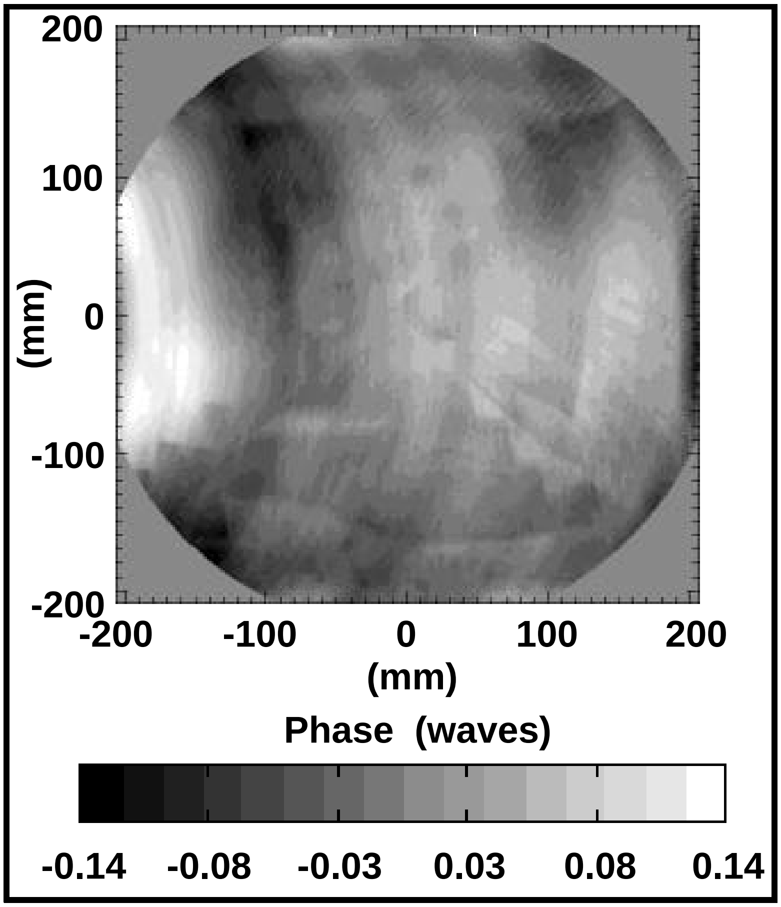

Fig. 8. Profile of wavefront distortion in a 40 cm circular section of the 41 x 41 cm^2 plate described in Fig. 7. Again, although sector and growth hillock boundaries are visible, the distortion is small.

7. ACKNOWLEDGMENTS

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