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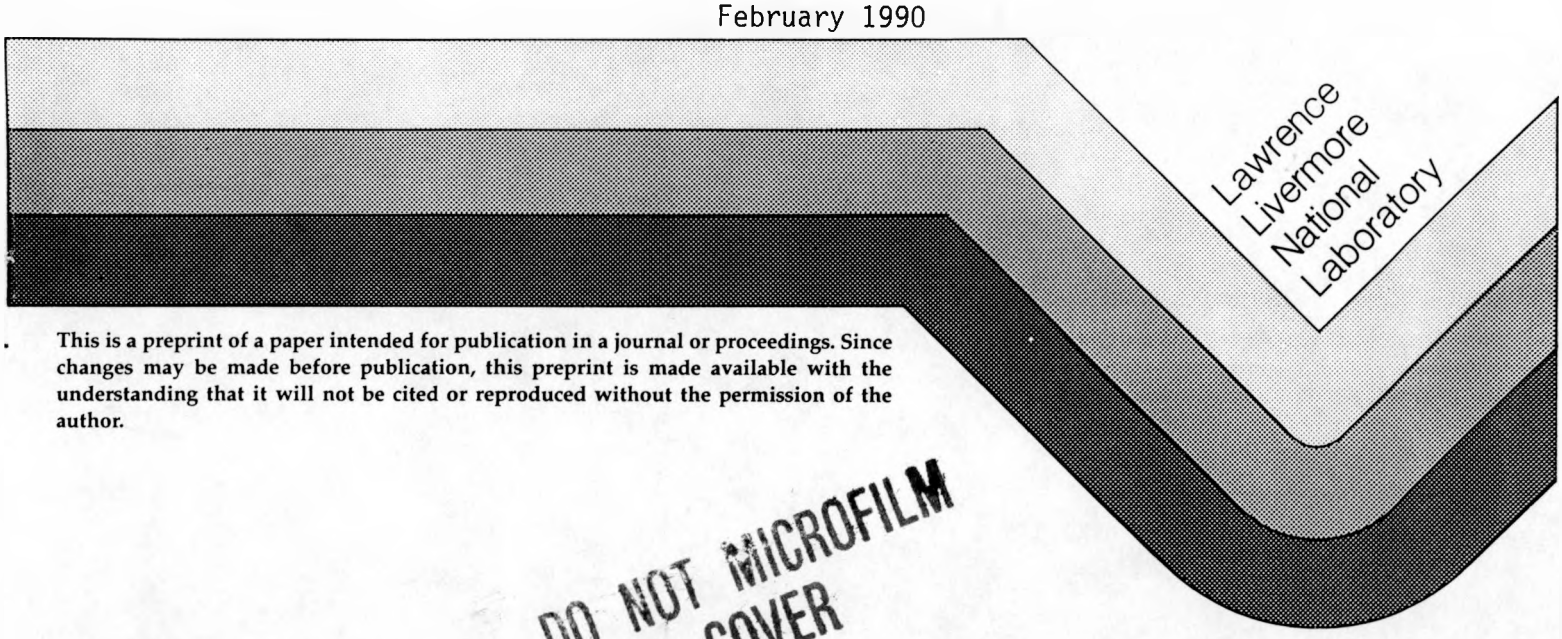

Development of a large scale Nd:YAG growth process

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ABSTRACT

Development of an automated steady-state process for Czochralski growth of flat interface Nd:YAG laser crystals is described. Steady-state is achieved through addition to the melt to maintain constant melt depth and composition. Interface flattening is accomplished through a controlled spin-up of crystal rotation during which characteristic changes in melt flow and crystal weight are observed. Combined steady-state/flat-interface growth has been demonstrated for sections up to 30 mm diameter x 110 mm long. In batch mode, without melt addition, flat interface YAG and Nd:YAG up to 52 mm x 220 mm, and undoped YAG up to 85 mm x 75 mm have been grown. In batch mode, gradual reduction of rotation rate is required to maintain interface contour and cross-sectional shape of the crystal as the melt height decreases.

1. INTRODUCTION

Neodymium-doped yttrium aluminum garnet (Nd:YAG) was reported to operate as a laser by Geusic, Marcos and Van Uitert in 1964.[1] Single crystals of Nd:YAG have now been available on a commercial scale for many years and are commonly fabricated into laser components in the shape of rods or slabs. Production of these crystals normally employs the Czochralski process of pulling from the melt, and the conventional implementation of this process involves growth (pulling) along a [111] crystallographic axis under conditions which produce a deep interface, i.e. the growing end of the crystal has an essentially conical shape, convex into the melt. This interface geometry, combined with {211} facets near the tip, generates a non-homogeneous central core[2] surrounded by a six-lobed strain pattern which limits the size of fabricated laser elements.

Incorporation of neodymium ions into the YAG lattice is governed by a distribution coefficient, k, generally agreed to have an effective value of about 0.2. This implies that at each stage of growth the crystal incorporates 20% of the neodymium concentration contained in the region of the melt which is immediately adjacent to the growing crystal. Since 80% of the neodymium is rejected, the concentrations in both the melt and the crystal build up as growth progresses. Growth of Nd:YAG crystals is usually limited to about 25% of the batch charge, in part to minimize the end-to-end gradient of neodymium concentration, but also to avoid the problem of constitutional supercooling which is aggravated by increasing concentration in the residual melt. The desire for large fabricated components therefore forces the use of large quantities of starting material and large crucibles in Czochralski growth of Nd:YAG using the batch process.

The batch process introduces complexity to the growth of a crystal through the progressive decrease in melt height. As the melt height decreases, transfer of heating power to the melt is effected and the strength of convective melt flow and mixing is reduced. The thermal environment of the crystal is also altered as the melt level falls, exposing the crystal to a greater area of the hot crucible wall. This tends to lower the temperature gradient in the melt, which enhances the risk of constitutional supercooling.

In order to circumvent some of the shortcomings of the standard process, development of an automated Czochralski growth process was initiated. Two key objectives were the achievement of steady state conditions and elimination of the central core through growth with a flat solid/liquid interface. The steady state condition was to be achieved through addition to the melt in order to maintain constant melt height and constant concentration of dopant in the melt. The normally deep interface was to be flattened through a programmed increase in rotation rate.

2. APPARATUS AND METHOD

2.1. Automated growth system

The basic growth station is conventional in that it consists of an inductively heated iridium crucible and thermally insulating furnace structure within a water cooled belljar. The atmosphere within the belljar is maintained by a flow of dry nitrogen to which oxygen is added at a level of about 0.25 volume percent. These gases leave the belljar at the top through a small opening provided for entry of the seed and seed holder. Oxygen and moisture contents of the atmosphere are monitored with appropriate instrumentation. Pulling of the crystal is achieved by means of a geared-down variable speed D.C. motor which drives a vertical precision ballscrew. The ballscrew lifts a platform which supports an electronic balance from which the seed holder is suspended. A variable speed D.C. motor rotates the seed holder. The electronic balance has a capacity of 12 kg, and a resolution of 0.1 gram. The induction heating unit operates at 10 kHz with a maximum power rating of 50 kW. Line voltage for the entire system is regulated and backed up for short term power outages by an uninterruptable power supply. Cooling water for the induction power supply and belljar is controlled within about 1^0F.

Automation of the growth process is implemented through a Digital Equipment Corporation PDP-11/24 computer equipped with both serial and HPIB interfaces. These interfaces provide communication with the electronic balance, a dedicated analog controller which drives the induction heating unit, the lift and rotation motors, and the feed system which adds material to the melt. Multiple growth stations can be operated by the computer on a timeshared basis and archival data is collected from each station and stored on hard disk media for subsequent analysis.

Computer software has been developed which operates on the basis of a "growth plan" which provides a profile for each programmed parameter as growth progresses. The growth plan is written in tabular spreadsheet format in which each time-based segment , or block, provides endpoint values to be achieved by each parameter.

The growth control algorithm operates on the mass rate of growth, in grams per hour, determined by performing a linear regression on a moving queue of weight readings collected from the electronic balance. This measured mass rate is compared with the scheduled mass rate , and the error is operated upon by adaptive proportional-integral-derivative (PID) parameters to make adjustments in power level of the induction heating unit. Measurement of mass rate is inherently noisy and lags behind about 15-20 minutes in this system, but rather good, repeatable control of growth can be achieved. A further improvement in control is realized through the use of a "power predictor profile" embedded in the growth plan. This profile is developed and refined over several iterations of the growth plan using archival data from successive runs. Refinement of the power predictor allows reduction in the PID control parameters to a level which reduces the effect of noise in the mass rate upon furnace power, but still maintains good tracking of the growth plan.

2.2. Steady state process

The objective in the steady state process is to maintain constant composition and volume of the melt in order to grow a crystal of constant composition under essentially constant thermal conditions. The composition and volume factors were addressed by adding makeup material to the crucible during growth. Addition of material to the melt during Czochralski growth of KCl and CsI:Na[3] and silicon[4] has been reported, but we are not aware of prior use of the technique in growth of optical laser crystals.

In this work, addition to the melt takes the form either of small pellets added periodically or of a rod of material being pushed into the melt at a steady controlled rate. Ideally the composition of added material matches the composition desired in the growing crystal, and the mass rate of addition matches the mass growth rate of the crystal.

2.3. Flat interface growth

Growth with a flat solid/liquid interface to eliminate the axial core in Nd:YAG dates back to at least 1967 when Cockayne, Chesswas and Gasson[5] reported its use for small crystals. The technique is commonly used in growth of flat interface gadolinium gallium garnet (GGG) for use as substrate material for magnetic bubble films[6,7]. It thus seems surprising that flat interface growth of Nd:YAG is not common. The problem is indeed not trivial, at least in large Nd:YAG crystals. Union Carbide was reported to have abandoned work on flat interface YAG growth around 1972 due to problems with dislocations.[8] Development of finesse in the process is very time consuming and expensive due to the slow growth rate (~ 0.25 mm/h) required for doped YAG. Many of the experiments in this work were therefore conducted with undoped YAG to speed up the learning process, since undoped YAG can be grown much faster (~ 4-7 mm/h).

The deep convex interface found in YAG at slow rotation rates is the consequence of several factors. Radiation dominated heat transfer away from the solid/liquid interface through a transparent crystal tends to create a lower temperature at the center of the interface. The radially inward flow of melt at the surface, due to convective flow driven by the hot crucible wall, is cooled as it moves across the growing interface. Both of these factors contribute to shaping the depressed center of the freezing isotherm.

Modification of the shape of the isotherm/interface shape might be achieved to some extent by using baffles or an afterheater to alter heat flow through the crystal, but the resulting decrease in temperature gradient would increase further the risk of constitutional supercooling.[2] The most accessible control factor seems to be that of rotation rate. The rotating crystal acts as a centrifugal pump, and as its rotation rate is increased the outward flow of melt competes more strongly against the inward convective flow.

3. CRYSTAL GROWTH EXPERIMENTS

3.1. Steady state growth

Melt addition studies were performed on both YAG and Nd:YAG crystals of about 27 to 30 mm diameter grown from 76 x 76 mm iridium crucibles using lids with inside diameters of 51 mm.

The "carousel" pellet feed shown in Fig. 1 was used to drop pellets through an alumina tube into a tubular iridium reservoir which is suspended in the melt at the side of the crucible. The upper disk of the carousel was rotated slowly by a small stepping motor. Pellets fell from the upper disk as they passed over a single hole in the lower disk. Most of the experiments were performed using single crystal pellets cored out of YAG or Nd:YAG cutting scrap. Typical pellets were about 5 mm diameter x 7 mm long weighing about 0.25 gram each. In some cases ceramic pellets and single crystal rods were used as feed material. Rod-feed employed a simple screw mechanism which slowly pushed the rod into the melt. It is worth noting that rod feeding was not a continuous process, even though the rod was pushed at a constant rate. Since the rod must be pushed into the melt near the edge of the crucible where

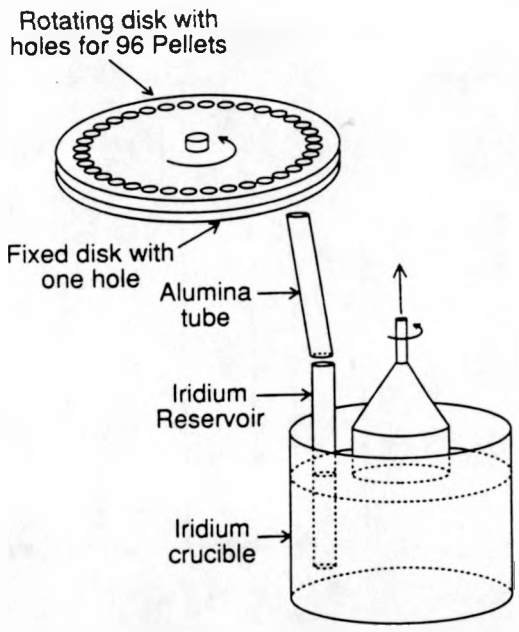


Fig.1 Carousel pellet feed.

maximum temperature prevails, the rod touches, melts off, re-touches, etc.

In early experiments with small 76 x 76 mm crucibles, longer pellets weighing about 0.5 gram each were tried. With that size pellets, changes in mass growth rate were observed which correlated with the dropping of each pellet. This effect was attributed to chilling of the melt by large pellets which were at room temperature. The effect of 0.25 gram pellets was undetectable within the normal noise level of the mass derivative. Regardless of pellet size, the growing crystal exhibits an increase to a new, stable diameter during addition to the melt. Elimination of the melt-fall component of linear growth rate requires an increase in diameter to sustain the same mass- (or volume-) growth rate.

Addition to the melt provided the conditions for steady state growth and dopant leveling, but it was soon apparent that the crystals contained many small particles of iridium. These inclusions started at the point where addition was started and gradually stopped if addition was discontinued. Careful drying of the pellets and use of a dry nitrogen atmosphere in the carousel unit did not help. Even without addition to the melt, iridium inclusions could be generated through relatively small changes in melt temperature, for example if the alumina pellet delivery tube was installed while growth was in progress.

The melt and iridium crucible are believed to reach an equilibrium in which the melt is saturated with iridium, probably in oxide form. Iridium nuclei which are present after initial melting of the charge drift around and grow until they settle to the bottom or become attached to the hot crucible wall during the conventional "aging" period before starting growth. New nuclei may be generated through chilling the melt, and these will then drift and grow until they either get trapped in the growing crystal or attach themselves to the crucible.

In one experiment a crystal was grown while adding pellets to a closed-end iridium reservoir so that there was no transfer of material from the reservoir to main melt. No iridium inclusions were produced in the crystal, indicating that the chilling effect of pellet addition did not extend out through the wall of the reservoir enough to generate new nuclei in the main melt. Generation of nuclei was apparently localized within the small volume ($\sim$5-10 cm^3) of the reservoir, so these nuclei needed to be prevented from reaching the main melt.

Fig.2 shows some of the reservoir designs which were tried in the effort to eliminate iridium inclusions. The bottom of the original open ended reservoir (5a) was closed, and small holes were made in the side (5b, 5c). The intent was to slow down the transfer of fluid and to provide a settling space for particles in the lower end, but this gave no improvement. Addition of a pre-melting shelf inside the reservoir above the level of the melt (5d) provided a substantial improvement. The small opening at the shelf prevented passage of added pellets until they melted, and thus reduced

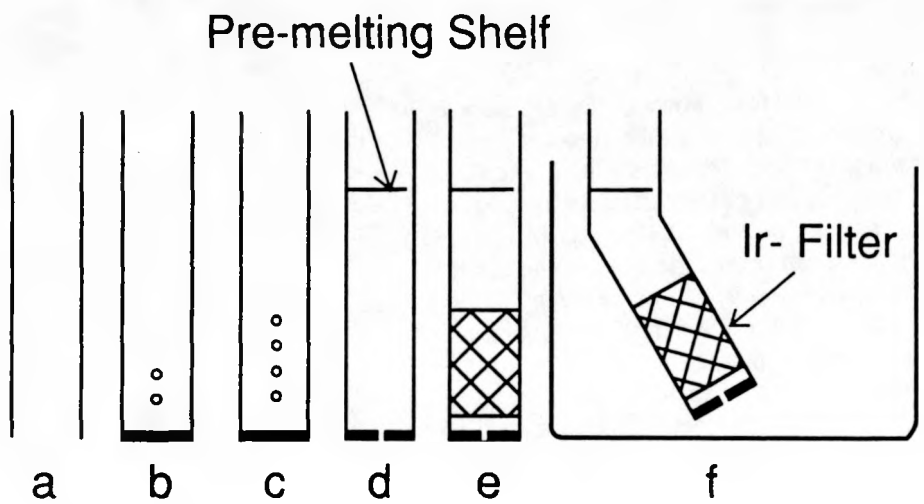


Fig.2 Reservoir designs.

thermal shock to the melt. Addition of a filter bed[10,11] of iridium particles below the shelf (5e) provided an additional safeguard. In the final design (5f), the shelf and filter were combined in an "L" shape reservoir which was mounted so that the new melt emerged over the center of the crucible floor. This has been observed to be a natural gathering spot for iridium particles and should therefore provide some additional help in trapping. Auxiliary cooling of the crucible floor has been used as a means of collecting iridium from the melt[9], but piles of iridium particles have been found in this area without resorting to supplemental cooling. The final design has completely eliminated the occurrence of iridium inclusion when using either single crystal or ceramic pellets.

3.2. Flat interface growth

All crystals in this study were grown using [111] orientation seeds.

Initial attempts at flat interface growth were programmed for 25-30 mm diameter Nd:YAG crystals pulled from a 76 x 76 mm iridium crucible with a lid having an inside diameter of 51 mm. Rotation rates were fixed, in the range of 40 to 65 RPM, such that a self-activated flattening "flip" would take place as the crystal approached full diameter.

In parallel experiments with 82 mm diameter Nd:YAG crystals, from 152 x 152 mm crucibles with 102 mm I.D. lids, several variants of rotation programming were tried. One plan called for flattening early in the cone, at about the diameter of the 30 mm crystals, then reducing rotation rate as the diameter increased. In another approach the rotation rate was held at 10 RPM until nearly full diameter was reached, then gradually increased to a rate which was predicted to achieve flattening. In both the small and large crystals the onset of melt pumping and reduction in mass rate of growth was observed, indicative of a transition in growth conditions. Examination of these crystals between crossed polarizers revealed that flattening did not occur as a singular event, but was spread out over some distance along the growth axis. The interface did not reach an essentially flat shape until the crystal had grown in length a distance roughly equal to one diameter past the initial observation of melt pumping. The transition process was apparently not smooth, since voids and dislocations were found, and continued growth resulted in cracking which apparently originated at these voids.

Lack of smoothness in the transition was probably due in part the attempt to maintain control over the mass rate of growth while encountering weight losses due to the changing shape of the interface. These losses were not clearly predictable with respect to time and rotation rate, so any attempt to build a program which properly handled these losses was more likely to exacerbate the control problem.

A different procedure was evolved in which the heater power was locked and the pull stopped, at a predetermined position in the boule, before increasing the rotation rate to initiated the flattening transition. The main objective was to provide reproducible thermal geometry in order to simplify modeling of the process, and to complete the transition before resuming pull. Since earlier crystals had flattened late, and usually well past the shoulder, the location of the transition was chosen to be about "one diameter" past the shoulder of the boule. This placed the shoulder at about the level of the crucible lid so that the radiation aperture between the crystal and the lid would be essentially constant for subsequent growth.

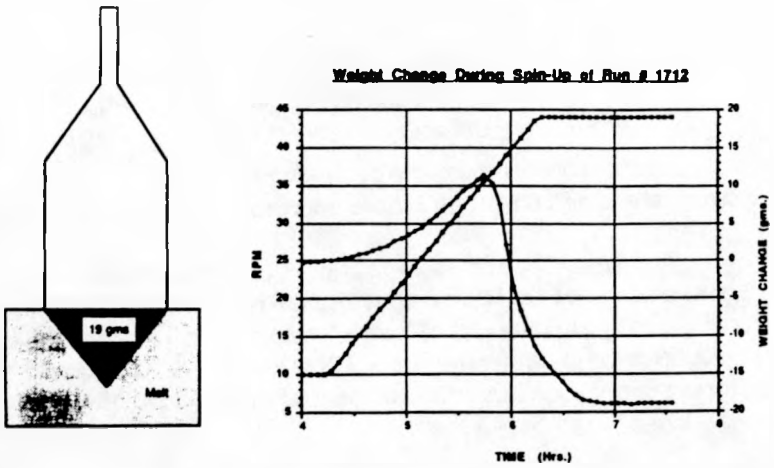


Fig.3 Rotation rate and weight changes during flattening transition.

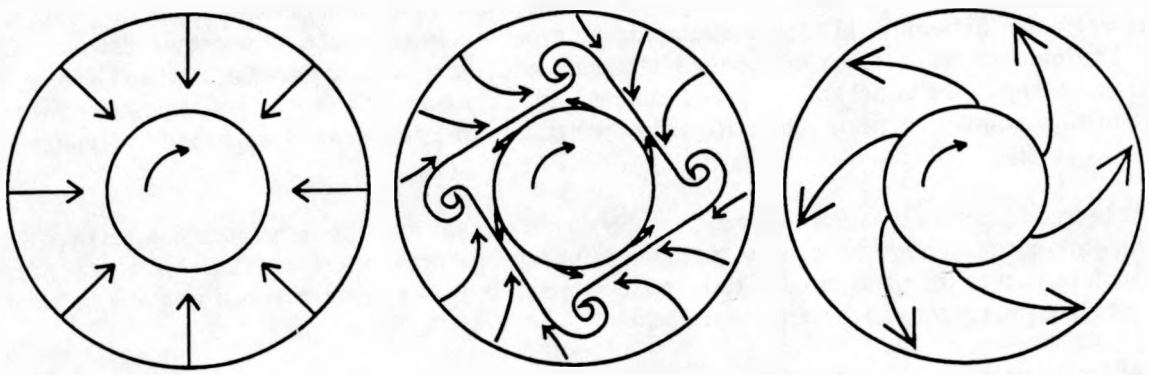
In order to monitor and analyze the flattening transition, the rotation rate was programmed to increase linearly over a period of several hours. During this spinup the compu-

ter automatically collected data at intervals of 3 to 5 minutes. Fig.3 is a plot of rotation rate and weight change from data taken during spinup of a 52 mm diameter undoped YAG crystal from 10 to 44 RPM. The crystal exhibits a gain in weight of about 12 grams, then starts to lose weight at about 36 to 37 RPM. Weight loss continues even after reaching the maximum rotation rate, but levels off with a net loss of 19 grams from start to finish.

Weight loss is equated with melting of submerged solid contained within the original deep interface.[12,13] The magnitude of the weight loss is determined by the volume of material melted and the difference in density between the solid and the melt. Assuming the solid and melt densities to be 4.56 and 4.01 g/cc, the 19 gram loss is equivalent to a right circular cone with a 52 mm diameter base and a 56 degree included angle. The same amount of weight loss may of course produce either a somewhat convex or concave final interface depending upon the shape of the original deep interface. It is therefore important to maintain a controlled growth environment in order to obtain reproducible initial and final interface shapes.

The weight gain before inception of the "flip" is not completely understood, but seems to represent actual addition of solid. Review of video tapes suggests that there may be a slight increase in diameter during this stage of spinup.

In addition to monitoring the weight change during spinup, the fluid flow at the surface of the melt was monitored visually and with a video camera recording on tape. Fig.4 summarizes these observations of the transition



Type I Type II Type III

Fig.4 Surface flow patterns observed during growth of YAG and Nd:YAG crystals from a 152 mm dia. crucible with an annular lid having an inside dia. of 76 mm.

from dominantly convective flow to pumped flow extending past the perimeter of the growing crystal. These observations are essentially in agreement with those reported by Brandle[14] and Whiffen[15], which were both based upon simulations.

The "spoke" pattern in Type I flow is associated with with the radially inward convective flow which dominates at low rotation rates (10 RPM), or with small crystal diameter. Type II flow represents the zone of outwardly pumped flow bounded by a variable set of "swirls", associated with essentially flat interface (~ 40-44 RPM for 52 mm dia.). Type III flow shows the dominantly pumped flow resulting from excessive rotation rates (50-60 RPM). This strong pumping produces a very concave interface shape, and three sides of the [111] axis crystal become progressively more hollow, or fluted.

In Type II flow, the spiral waves moving outward within the zone of pumped flow originate at the six corners of the boule crossection associated with growth along the [111] axis. In Type III flow these waves extend outward under the annular lid, and may reach the wall of the crucible. As the crossection of the boule becomes fluted, and more

triangular or "lobed", the outward flow pattern shifts to three heavy spiral waves. Continued growth with Type III flow often leads to "dewetting" in which the crystal loses contact with the melt.

The marked visibility of surface flow patterns in YAG (and GGG) melts is believed to be representative of liquids having high temperature coefficients of expansion. In contrast, the nearly featureless surfaces of alexandrite and lanthanum beryllate melts are believed to result from low temperature coefficients and/or low radial temperature gradients.

After spinup the weight was allowed to stabilize before resuming PID control and pull. Maintenance of a stable interface shape during growth after spinup presented some difficulty. In some cases the interface flipped from flat or slightly concave to slightly convex. This generally introduced voids, due to rapid growth in the center, and these voids appear to be the source of cracking during a later stage of growth. Cracking was frequently catastrophic, resulting in dewetting of the crystal from the melt and even remelting the end of the crystal below the crack(s). Cracks act as barriers to both radiant and conductive heat transfer, causing the lower end of the crystal to become superheated. Less severe cracks were handled well enough by the mass rate control system so that growth continued, but in these cases the power level was reduced by control action to accommodate the diminished heat transfer.

Growth with a slightly concave interface has been maintained for lengths of more than 200 mm at 52 mm diameter, but three sides of the boule became progressively more fluted (concave) to the extent that the end of the boule looks like a three-lobed-end-mill. This can happen even when the initial interface is flat, following the transition, unless the rotation rate is gradually reduced at an appropriate rate as the melt depth decreases.

Nikolov et al[16,17] conducted extensive simulations of fluid dynamics in Czochralski growth using glycerol-water solutions and even used interactive interface structures made of a mixture of paraffins so that the shape actually changed as a the flow changed. They concluded, in part, that the critical rotation rate for flattening is proportional to $h^{0.155}$, where h is the height of the melt.

Fig.5 shows a longitudinal section of a 52 mm diameter Nd:YAG crystal as viewed between crossed polarizers. No material was added to the melt during growth. The core is clearly visible in the upper section, which was grown at a rotation rate of 10 RPM. The deep convex interface appears as very faint lines in the original photograph, delineating a nearly conical shape with an included angle of about 65 degrees. The core was terminated by stopping the pull and "spinning up" the rotation from 10 to 44 RPM over a two hour period. The resulting new interface is essentially flat with a slightly wavy "m" shaped contour. Pulling was resumed, and continued for seven days while slowly reducing the rotation rate (proportional to $h^{0.15}$) to 43.5 RPM. The decanted terminal interface is approximately 2 mm concave.

Growth of longer sections of undoped YAG without melt addition and with rotation "tapers" proportional to $h^{0.15}$ produced a progressively more concave interface, and a fluted crossection. In several cases the crystals dewetted, losing contact with the melt.

Subsequent growth runs without melt addition used rotation tapers with proportionalities rang-

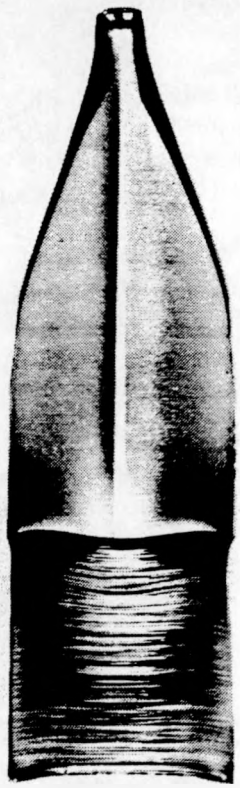


Fig.5 Section of a short 52 mm diameter Nd:YAG between crossed polarizers showing the termination of the core at the flattening transition.

ing from $h^{0.25}$ to $h^{0.35}$. Core-free sections up 52 mm diameter x 220 mm long were produced with at most slight changes in their cross-sectional shapes even though the terminal interfaces were up to 5 mm concave.

The central core and associated strain pattern have been eliminated, but at this stage, the core-free sections of neodymium doped boules contain bands of fine bubbles along the central region. These bands place some restriction on cutting slabs, but far less restriction than is imposed by the usual core and strain pattern. Several slabs approximately 5 x 35 x 210 mm could be fabricated from each of the above core-free sections. These sections have doping levels of 0.5 x 10^{20} Nd ions per cubic centimeter.

No specific cause for the fine bubble bands in the doped crystals has been identified at this time. The bubbles, or voids, may be the result of constitutional supercooling, which would indicate the need for slower growth rates. These boules were pulled at 0.25 mm/hour, requiring close to two months for the full growth cycle. Slower growth rates are not to be considered lightly, since the probability of equipment and utility problems would be increased, but the potential yield of large components or the increase in yield of smaller components might provide the justification.

4. CONCLUSIONS

An automated Czochralski growth system has been developed which is quite versatile in its ability to program and control many parameters of the growth process. The mass-rate control algorithm has been used for mass rates of growth ranging from less than 0.1 gram/hour to over 160 gram/hour with excellent control.

The steady-state process of dopant-leveling has been demonstrated in growth of core-free Nd:YAG crystals up to 30 mm diameter by 110 mm long. Iridium inclusions have been eliminated by modifications made to the pellet delivery reservoir.

Controlled flat-interface growth, without addition to the melt, has produced useful core-free sections of YAG and Nd:YAG up to 52 mm diameter x 220 mm long. Several slabs of about 5 x 35 x 210 mm can be fabricated from each of several doped sections of this size. An 85 mm diameter x 75 mm long core-free section of undoped YAG has been grown, its length limited by impending shutdown of building power for major maintenance.

Bands of fine bubbles or voids found at present along the center of core-free Nd:YAG sections cause much less restriction on fabrication of large sections than the usual core and strain pattern. Slower growth rates may be required to eliminate these defects.

5. ACKNOWLEDGMENTS

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