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A Comparative Study of the Influence of Buoyancy Driven Fluid Flow on GaAs Crystal Growth*

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ABSTRACT

This study consists of a systematic investigation of the effect of gravity-driven fluid flow on GaAs crystal growth, and will include GaAs crystal growth in the microgravity environment aboard the Space Shuttle. The program involves a controlled comparative study of crystal growth under a variety of earth-based conditions with variable orientation and applied magnetic field in addition to the microgravity growth. Earth-based growth will be performed under stabilizing as well as destabilizing temperature gradients. The boules grown in space and on earth will be fully characterized to correlate the degree of convection with the distribution of impurities. Both macro- and micro-segregation will be determined.

The space growth experiment will be flown in a self-contained payload container through NASA's Get Away Special program. The advantages of using the GAS program are simplicity of manifesting the payload aboard the orbiter, frequent flight opportunities, quick turnaround necessary for iterative experiments, and low cost. The payload with its large alkaline battery power source will include two redundant experimental systems including separate well insulated growth furnaces. Each sequentially scheduled growth experiment will require approximately 8 hours to complete, and collected data will include micro acceleration, temperature, and furnace power. The use of the specially designed growth ampoule and furnace system for both space- and earth-based growth experiments will lend validity to the comparative studies and simplify the numerical modeling.

1. INTRODUCTION

During the last decade, GaAs has become one of the most important electronic materials. GaAs exhibits an intrinsic electron mobility greater than 9000 cm^2/V-s making it a desirable material for very high speed signal processing devices, and its direct energy gap makes it a useful material for light emitting devices. Recently, undoped semi-insulating GaAs substrates of good quality have become available due to improvements in Liquid Encapsulated Czochralski (LEC) growth technology. The availability of state-of-the-art substrate material

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has given increased impetus to GaAs integrated circuit technology. Nonetheless, the current GaAs IC yields remain very low. It is not clear to what extent this yield problem should be ascribed to the substrate quality or to the still infant IC processing technology. However, it is clear that in the long run, GaAs IC technology will be substrate-limited unless substrate quality is improved.

While there has been much progress lately in increasing the yield of semi-insulating ingots with attractive transport properties, there has been only moderate success in lowering the dislocation density or enhancing the homogeneity of LEC grown material. Dislocation densities in LEC material are typically $> 10^4 \mathrm{cm}^{-2}$. Several groups have shown that the incorporation of an isoelectronic dopant, such as In, into the GaAs lattice can reduce the dislocation density to $< 10^3 \mathrm{cm}^{-2}$ with no appreciable degradation of the transport properties. Significant reductions in dislocation density have also been achieved through careful tailoring of the thermal environment in the furnace during growth. Even with these very recent improvements, the dislocation density in LEC GaAs remains high relative to Si. In addition, the resistivity and mobility in most GaAs boules varies both radially and axially, and pronounced growth striations are a clear indication of the inhomogeneous distribution of impurities throughout the ingot.

A major cause of these inhomogeneities is the significant degree of temperature fluctuation at the growth interface. In a gravitational field, density gradients in the melt caused by temperature gradients or compositional variations induce appreciable convection currents which produce pronounced turbulence at the growth interface and results in erratic temperature fluctuations. Such temperature instabilities are particularly unfavorable for the growth of III-V compounds such as GaAs and InP in which one of the components is quite volatile. This condition can result in constitutional supercooling and can require a higher temperature gradient for proper crystal growth. In addition, the convection induced temperature fluctuations introduce a fluctuating microscopic growth rate that leads to the inhomogeneous distribution of impurities.

Crystal growth of semiconducting crystals in space has clearly demonstrated that the elimination of convection-driven turbulence at the growth interface can substantially improve crystal quality. Examination of doped crystals of InSb(1) and Ge(2) grown in the microgravity environment of space yielded no evidence of growth striations. In contrast, impurity striations arising from an inhomogeneous incorporation of impurities during growth are invariably present in earth-based Czochralski grown crystals indicating an inhomogeneous incorporation of impurities during growth. In general, segregation in crystals grown under microgravity conditions is primarily diffusion-controlled. This mechanism results in a more uniform distribution of impurities or dopants along the axis of the crystal.

Convection currents in the melt can also be suppressed by the application of a magnetic field during growth. The magnetic field can be viewed as increasing the effective kinetic viscosity of the melt by resisting fluid motion through Lenz's Law. Several laboratories, including our own, have been investigating the utility of magnetic field assisted growth in improving the homogeneity of GaAs.(3-7) Results at GTE Laboratories,(3) illustrated in Figure 1, have shown that, for boules grown in a transverse field > 2000 G, erratic growth striations can be totally eliminated. Moreover, the field-grown GaAs exhibits dislocation densities of $< 10^3 cm^{-2}$. This decrease in dislocation density is probably due to the stabilization of the microscopic growth rate by the magnetic field.

Space-growth and magnetic field growth results demonstrate that convection can profoundly influence the quality and properties of semiconductor crystals. It is important to determine to what degree convection-induced temperature instability at the growth interface affects impurity segregation and development of defects in the crystal. A comparison of the effectiveness of magnetic suppression of convection to the elimination of buoyancy driven flow under microgravity can lead to improved earth-based growth of high quality GaAs under applied magnetic field and optimized temperature gradients. Enhancing the homogeneity and reducing the dislocation density of GaAs are two of the major goals of current research by the GaAs growth community.

2. SCOPE OF EXPERIMENT

In this study, a series of experiments has been designed to delineate the role of buoyancy driven fluid flow on the growth of GaAs single crystals under a variety of conditions. The approach chosen for this study is a comparative one in which microgravity growth in space serves as the limiting case of negligible buoyancy driven convection. The microgravity growth experiments are complemented by a series of earth-bound growth experiments that examine the influence of convection on GaAs growth as a function of the orientation of the temperature gradient and the presence or absence of a magnetic field.

The earth-based growth series includes field-free and field-assisted growth. Comparison of the flow-dampening effects of transverse and axial magnetic fields are included in the study. The experiments also include an investigation of the temperature gradient encountered in the three common modes of bulk crystal growth: horizontal Bridgman (HB), vertical Bridgman (VB) and Czochralski (Cz). The various growth modes are illustrated schematically in Figure 2.

The heart of the experimental apparatus to be utilized for the studies is a specially designed furnace and growth ampoule, two of which are mounted in a payload designed for inclusion in one of NASA's Get Away Special payload containers for flight in the cargo bay of the Space Shuttle. The payload is illustrated in Figure 3 and is similar to one successfully operated by GTE Laboratories on STS-41B in February 1984, for studying convective effects in arc lamps.

3. EXPERIMENTAL DETAILS

This experiment was designed to be flown in the GAS program because of the unique opportunity it affords for performing sophisticated experiments using the simple, inexpensive, and timely procedures it provides. This program offers the extensive advantages of fast processing, streamlined safety reviews, and flight opportunities on virtually every Shuttle flight.

The payload is 20 inches in diameter, 26 inches high and is suspended from a top plate provided by NASA. The growth furnaces are contained within two 6 inches in diameter, 12 inches long cylinders mounted side by side near the center of the payload. Since no power is available from the orbiter, large battery packs above and below the furnaces supply power for the growth runs. Two isolated microprocessors will respectively control the 2 experiments which will be performed at different times. They will also store in non-volatile EPROM memory 16 channels of data including furnace power, battery voltage, various temperatures, and 3 axes of acceleration.

A diagram of the growth ampoule and furnace is shown in Figure 4. The growth ampoule will contain a pregrown single crystal ingot of GaAs 4 inches long by 1 inch in diameter. The ampoule is fitted with a spring loaded plunger to maintain the geometric integrity of the charge thereby ensuring that there will be no free fluid surfaces in the growth compartment. Under these conditions, Marangoni convection will be eliminated in both earth-based and microgravity experiments. This is a double-walled ampoule especially designed to permit GaAs growth in a spring-loaded system. In this configuration slight pressure-induced distortion of the outer quartz wall will not inhibit piston movement. The growth ampoule will be provided with a means of growth interface demarcation to permit a determination of growth rate as well as the shape of the growth interface. The design will permit earth-based growth in the three ampoule orientations illustrated in Figure 2, corresponding to the three growth modes. Thus, the growth geometry will be invariant throughout the course of the space and earth-based growth experiments, making possible the comparative study.

The furnace surrounding the growth ampoule will be used in both space and earth-based growth to ensure that the details of the temperature gradient are well controlled. The furnace will accept an axial field solenoid of reasonable size, and it is compatible with the transverse pole gap of the existing electromagnet at GTE Laboratories. Thus, the growth geometry can be held constant during the axial and transverse field-assisted growth series as well. The platinum-wound, Zircar®-insulated furnace is capable of operating at 1200-1300°C at a power level of about 130 watts. The furnace is wound to provide a temperature gradient of about 20°C/in at the growth interface.

In a typical run, the furnace is equilibrated at a temperature that provides melt-back of 3 inches of the pregrown ingot. The remaining 1 inch of unmelted ingot serves as a seed for the regrowth experiment. The furnace is then cooled at a controlled rate to provide a regrowth-rate of about .75 in/hr. Total duration of a run is 8 hours — 2½ hours for warm-up and equilibration, 4 hours for regrowth and 1½ hour for controlled cool-down. Figure 5 is a power consumption vs. time plot of one such experimental run where the solid line represents the steady state power requirement. Area A represents the excess power required for melt-back and area B represents an equivalent decrease in heat input during solidification. The well-behaved power curve at section B indicates a fairly uniform growth rate. For the Space Shuttle portion of this study two sequential runs will be performed in the two independent GAS payload furnaces.

This study is directed primarily toward the determination of the effect of buoyancy driven fluid flow on axial and radial segregation of impurities. The pregrown ingots will all be doped with Se at a concentration range of 2 to $6 \times 10^{17}/\text{cm}^3$. Selenium was chosen as the dopant because its segregation coefficient is a constant in this concentration range and because it is a shallow donor whose concentration at a particular point can be determined by IR absorption methods.

The regrown ingots will be sectioned as shown in Figure 6. Two axial slices will be taken from each ingot. The remaining portion of the ingot will be diced transversly to provide semi-circular wafers of each portion of the ingot. The axial slices can be profiled to give detailed information on both axial and radial segregation. This can be achieved by both SIMMS and IR absorption methods. Since interface demarcation will be used on half of the runs, IR microscopy will be used on the appropriate axial slices to determine both micro- and macroscopic growth rates as well as interface shape. The semicircular wafers will be examined to determine the effect of growth parameters on defect density and distribution. Dislocation density and distribution on these wafers will be determined by microscopic examination of wafers etched in molten KOH. The wafers will also be examined by cathodoluminescence techniques to further characterize the nature and distribution of dislocations and other defects.

Several experimental runs have been made with the apparatus described above to optimize the temperature gradient and other growth parameters. Figure 7 is an infra-red micrograph (approximately 200X) taken from an axial slice of one of the regrown boules. This boule was grown in the VB configuration. The photograph clearly shows the initial regrowth boundary with the growth striations in the original LEC grown portion below and the absence of striations in the upper portion regrown in the stabilizing gradient of the VB configuration.

Numerical analysis of the growth furnace and the GaAs crystal growth in various orientations is now in progress. A significant part of this analysis will be performed in collaboration with Prof. R.A. Brown of MIT.

The series of earth-based growth experiments described is now in progress and the microgravity growth runs will proceed when the Space Shuttle flights are resumed.

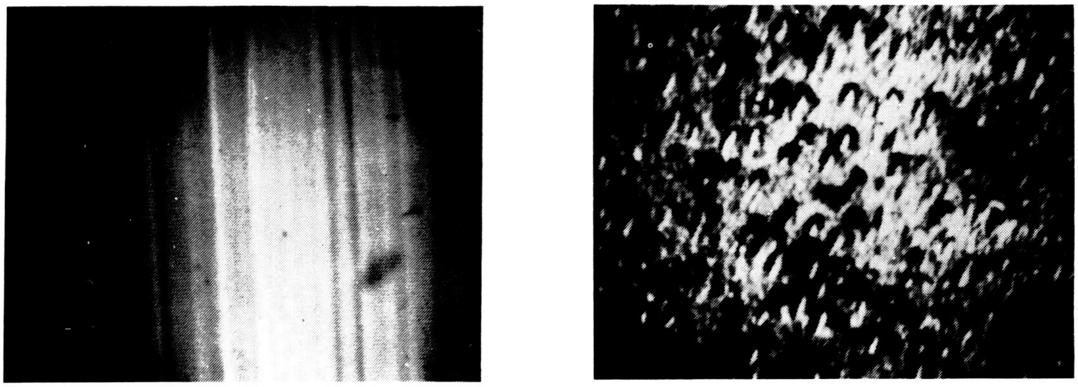
This comparative study should increase our understanding of the effect of convective flow on crystal growth in general and of GaAs growth in particular. The inhomogeneous impurity distribution and high defect density is a matter of great concern to the GaAs integrated circuits and opto-electronic device industries. We have already shown that magnetic damping of convective flow improves the homogeneity and reduces the defect density of earth-grown GaAs crystals. A direct comparison of the effect of free convective flow to magnetic damping and to the stabilization of the instantaneous growth rate under microgravity should prove valuable.

REFERENCES

1. A.F. Witt, H.C. Gatos, M. Lichtensteiger, M.C. Lavine, and C.J. Herman, J. Electrochem. Soc., 122, 276 (1975).
2. A.F. Witt, H.C. Gatos, M. Lichtensteiger, C.J. Herman, J. Electrochem. Soc., 125, 1832 (1978).
3. J.A. Kafalas in ″Gallium Arsenide Technology,″ David K. Ferry, Editor, pp 70-72, Howard W. Sams & Co. (1985).
4. K. Terashima, T. Katsumata, F. Orito, T. Kikuta and T. Fukuda, Jap. J. Appl. Phys., 22, L325 (1983).
5. K. Terashima and T. Fukuda, J. Cryst. Growth, 63, 423 (1983).
6. T. Fukuda, K. Terashima, T. Katsumata, F. Orito, S. Ozawa, T. Obokata and T. Sato, paper presented at Sixth American Conference on Crystal Growth, Atlantic City, NJ, July 1984.
7. J. Osaka and K. Hoshikawa, paper presented at Third Conference on Semi-Insulating III-V Materials, Kah-nee-tah, Warm Springs, OR, April 1984.

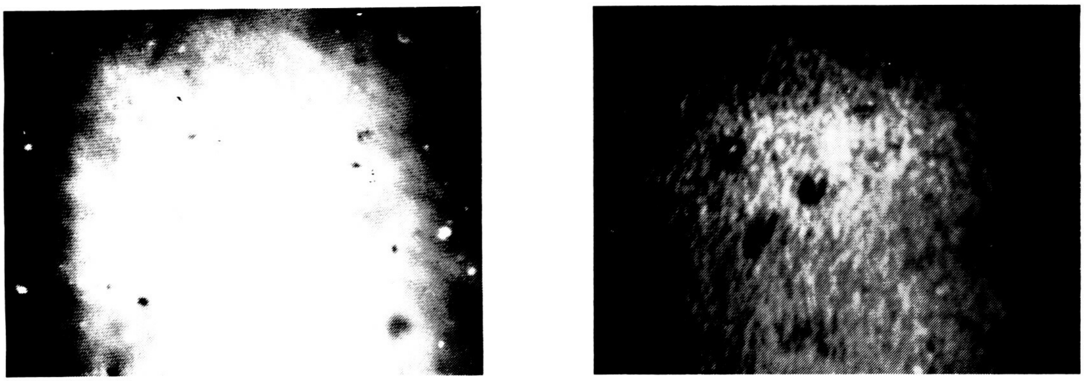
BIBLIOGRAPHIES

1. ″A Comparative Study of the Effect of Bouyancy-Driven Fluid Flow on GaAs Crystal Growth,″ J.A. Kafalas and A.H. Bellows, 6th Eur. Symp. on Mater. Sci. Under Microgravity, Bordeaux, France (1986).
2. ″The Use of Selective Pair Luminescence to Characterize Semi-Insulating GaAs,″ J.A. Kafalas, E.S. Koteles, S. Zemon and P. Norris, 13th Int. Symp. on GaAs and Related Compd., Las Vegas (1986).
3. ″Stability of the EL2 Center in GaAs Under Electron-Hole Recombination Conditions,″ J.A. Kafalas, M. Levinson and C.D. Coombs, Phys. Rev. B34, p. 4358 (1986).
4. ″Configurational Site Symmetries of the EL2 Center in GaAs,″ J.A. Kafalas and M. Levinson, to be published.
5. ″The Comparative Study of the Influence of Convection on Gallium Arsenide Growth,″ J. Gustafson, A. Bellows and J. Kafalas, Proc. of 2nd Pathways to Space Exp. Workshop, Orlando, FL (June 1986).



Striations at zero field

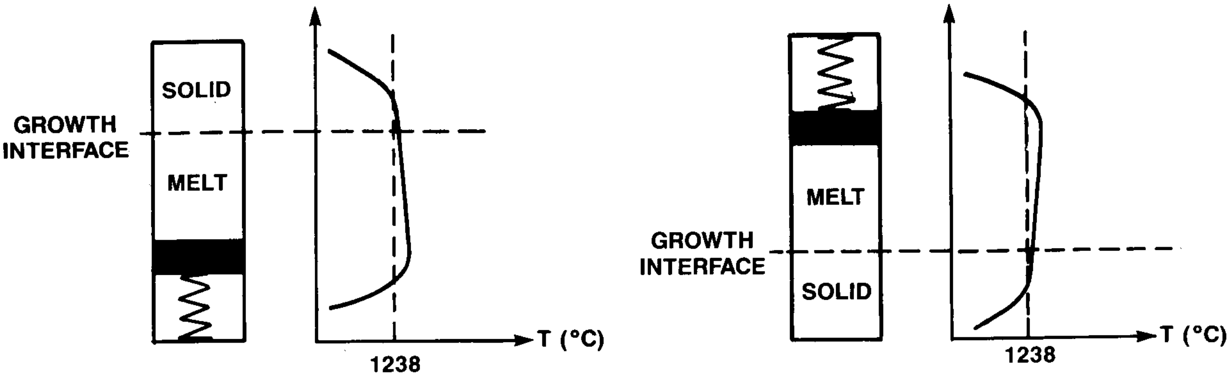
EDP at zero field



Absence of striations at 2000 G

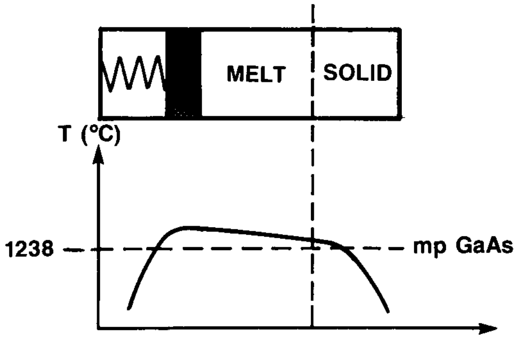
EPD at 2000 G

Figure 1. Comparison of striations and Etch Pit Densities in "field-off" and "field-on" regions of a crystal.



Czochrolski Mode (Cz)

Vertical Bridgman Mode (VB)



Horizontal Bridgman Mode (HB)

Figure 2. Schematic of the 3 different terrestrial growth orientations with the corresponding temperature gradients.

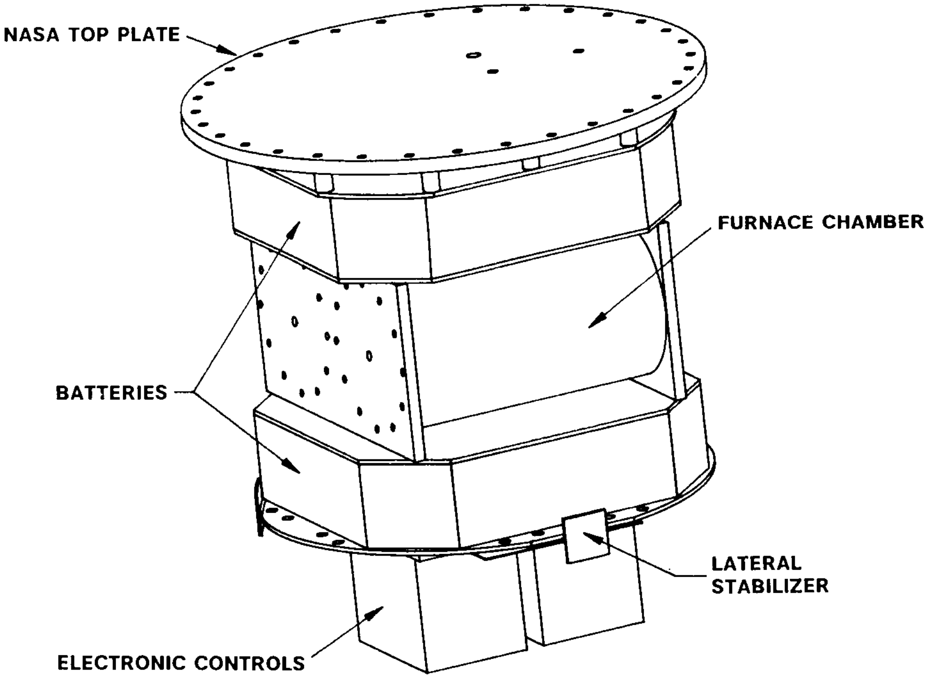


Figure 3. GAS payload for GaAs crystal growth.

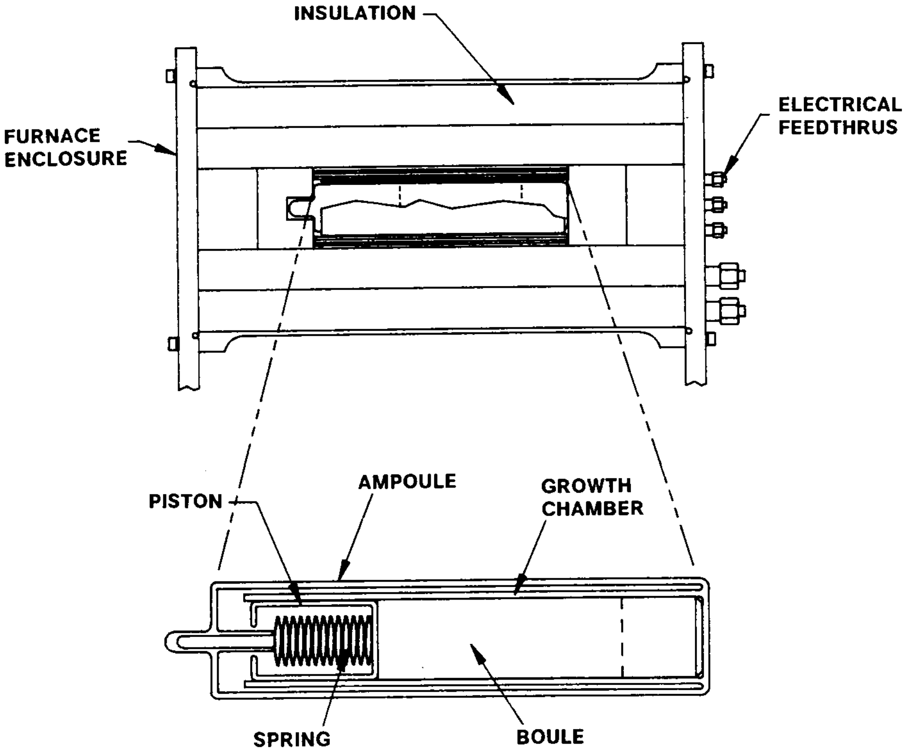


Figure 4. Section view of cylindrical growth furnace and enlarged section of double walled growth ampoule.

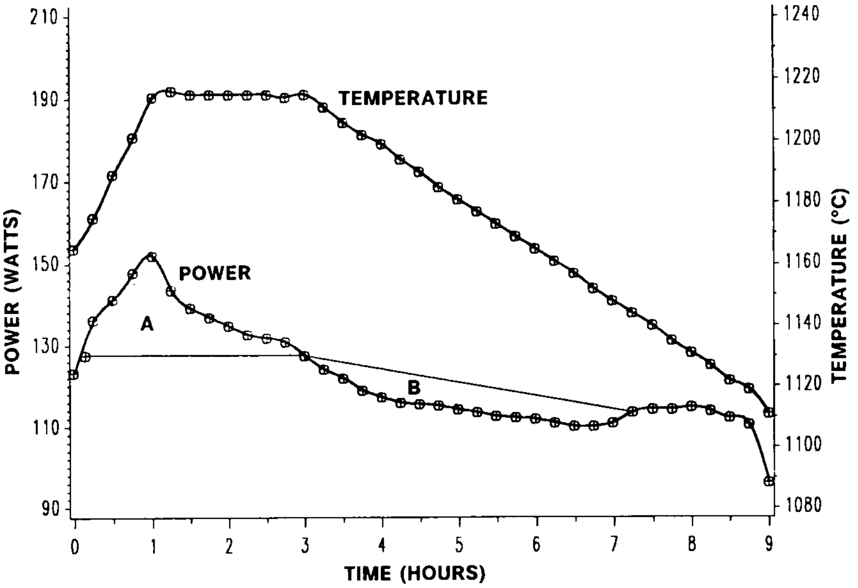


Figure 5. Power vs Time for a typical growth run showing incremental power required to melt boule and equivalent power retrieved during regrowth.

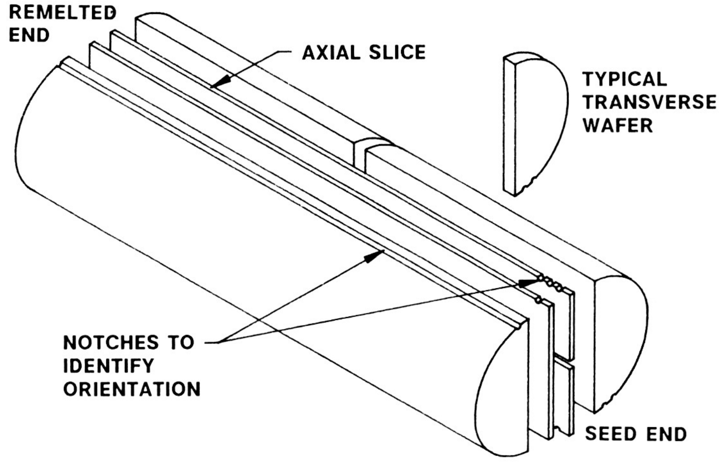


Figure 6. Dicing scheme showing pattern of notches used to permanently indicate orientation of each die.

Figure 7. IR micrograph showing growth striations below the regrowth boundary and evidence of uniform growth above the boundary.