Crystal growth from melt in space*

P GANGULY

Solid State and Structural Chemistry Unit, Indian Institute of Science,
Bangalore 560 012, India

MS received 21 June 1980

Abstract. A review of experiments on crystal growth from melt in nearly zero gravity environments in space is given. The review includes experiments from Skylab and ASTP missions. The results discussed are morphological observations, melt interface observations, dopant segregation, alloy solidification and containerless experiments.

Keywords. Crystal growth, zero gravity.

1. Introduction

In this era of laser and bubble domain memory devices the necessity for perfect crystals for use in what has been termed the opto-acoustico-magneto-electronic solid state devices industry has been well recognised. Table 1 lists examples of materials required for various applications in industry. The achievements of the crystal growers with their feet solidly on the ground have been truly stupendous. In table 2, a list of compounds grown from the melt by the Czochralski method is given showing that almost all range of melting points is now available for crystal growth. Indeed, in an analysis of the future of crystal growth towards the year 2000, Laudise (1974) does not find much need to comment about growth of crystals in space.

Table 1. Crystal growth and industry.

Application	Examples of materials used
Solid state laser	$\text{Y}_3\text{Al}_5\text{O}_{12}\text{-Nd}^{3+}$
Insulating substrates	Al_2O_3
Substrates for magnetic materials	$\text{Gd}_3\text{Ga}_5\text{O}_{12}$
Surface acoustic wave devices	LiNbO_3
Bulk wave devices	MgAl_2O_4 , LiTaO_3
Ionic conductors	$\beta\text{-Al}_2\text{O}_3$, Na_2WO_3
Non-linear optical devices	LiNbO_3
Optical windows	Al_2O_3 , CaF_2 , LiF
Acoustic optic devices	PbMoO_4
Optical polarizers	CaCO_3 , DyVO_4 , NaNO_2
Magnetic semiconductors	Fe_{1-x}S ($0 < x < 0.1$)

* Communication No. 77 from the Solid State and Structural Chemistry Unit.

Table 2. Crystals grown by Czochralski method.

Crystal	Melting points (° C)
NaNO ₂	271
Pb ₅ Ge ₃ O ₁₁	738
LiYF ₄	838
PbMoO ₄	1050
CaWO ₄	1570
Y ₃ Al ₅ O ₁₂	1970
MgAl ₂ O ₄	2105
Y ₂ O ₃	2410

The state of the art is, however, far from perfect. The advent of technology which has improved the technique of crystal growth has also increased the capability of the scientists to look at the defects in the crystals in greater detail and thereby understand better the mechanism of crystal growth in a truly dialectical fashion. Some of the types of defects encountered are shown in figure 1. Nevertheless, the growth of crystal, 5 to 10 cm in diameter and several feet long, is common place and sometimes one has to worry more about seed support and deformation of crystals under their own weight.

In order to control the position, shape and local morphology as well as to prevent uncontrolled nucleation, a temperature gradient is generally maintained at the freezing interface during the growth of crystal from the melt. Whenever the temperature gradient is not aligned parallel to gravity, convective flow arises. Similar gravity-driven convection arises whenever concentration gradients give rise to density differences. At low values of the applied horizontal temperature gradient the basic convective flow is steady. At higher temperature gradients, transition to time-dependent flows takes place and turbulence is eventually reached. The conditions for convective flow have been described in great detail elsewhere in this conference. The critical number is the Rayleigh number $N_R = N_{GR} N_{PR}$ where N_R is a measure of the temperature gradient, N_{GR} being the Grashoff number which is a measure of the horizontal temperature difference and N_{PR} is the Prandtl number which is the ratio of momentum transfer to thermal transfer.

The principal effect of convection is to produce a fluctuating temperature at the crystal-melt interface which results in an irregular growth rate. Gravity-driven convection may be eliminated by orienting the temperature gradient with respect to the gravitational field so that the dense fluid is at the bottom especially when the species rejected into the melt from the interface have greater density than the melt. Irregularities due to convective flow could also be corrected for rather dramatically in conducting samples by the applications of a small external magnetic field. The purpose of the space experiments was to eliminate the effect of gravity-driven convection in the near zero gravity conditions (10^{-4} to 10^{-6} g) of

Figure 1. Typical defects in crystals (a) scanning electron micrograph of voids in Czochralski single crystals of CaWO_4 showing internal structure (figure 2 of Cockayne 1977). (b) SEM of voids without internal structure in Stockbarger single crystals of LiRF_4 . (c) Transmission electron micrograph (TEM) of low angle tilt boundary observed in Czochralski single crystals of YAlO_3 (bar marker = $0.75 \mu\text{m}$). (d) TEM of inversion boundaries in Czochralski single crystals of LiTaO_3 . (e) X-ray reflection topograph of Czochralski single crystals of $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ showing interface shape, growth striations, dislocation facets (bar marker = 2 mm).

an orbiting space station. It should be remembered, however, that although zero-gravity environments could substantially reduce gravity-driven convection effects, there are other effects such as surface tension gradient-driven thermocapillary or Marangoni flows, flows arising from pressure gradients due to chemical reactions, thermoacoustic convection, low level g-jitter, or phase-change-driven convection especially when there is a large volume difference between the melt and the solid. Besides the above flows, the effect of variation in other fluid properties such as viscosity, thermal conductivity and thermal expansion, due to a temperature gradient has not been studied in the context of crystal growth from melt. Moreover, the beneficial effects of one-g conditions such as achievement of uniform composition by turbulent convective mixing and the elimination of unwanted bubbles will be absent in space.

There are intrinsic processes also which contribute to the formation of defects and the variation of morphology of a crystal. In a crystal one actually observes growth forms and not equilibrium forms so that isotropic diffusion effects cannot account for the variation in morphology. As is well-known the growth rate of the crystals differs for different faces of the crystal leading to the development of facets which may control the magnitude and anisotropy of the crystallographic aspects of the growing crystal. Secondly, in the growth of crystals of binary mixtures from the melt, the freezing temperature of the liquid increases away from the interface, as the concentration of the solute in the melt is greater at the interface than at the bulk. There is therefore the possibility of what is known as constitutional supercooling near the interface, especially when the temperature gradient at the interface is not too steep. This could lead to cellular growth in metals provided faceted growth does not lead to stable faces despite a super-cooled medium. Gravity-free environments cannot prevent the formation of defects due to the above processes. Instead one may argue that a convective mass flow could even out to a certain extent the adverse consequences of constitutional supercooling. We present in this paper a brief review of the experiments in space connected with the growth of crystals from melt.

Experimental

In the actual experiments that were carried out in the *Skylab III*, *Skylab IV*, and the *Apollo Soyuz Test Programme (ASTP)* missions, the experimenters faced certain limitations, as will be pointed out later, such as low electrical power, restricted ways of handling samples, etc. Moreover, because of the high cost of each experiment reproducibility was not tested for. On hindsight, at least, one gets the impression that there was not a proper appreciation of the problems which could have been anticipated and avoided. Initial reports of spectacular results were perhaps due to some sort of wish fulfilment. This was replaced subsequently by serious thoughts on the need for careful preparation of the experiments, and the need for further earth-based experiments before these experiments were carried out in space.

The scope of the present paper does not allow a detailed report on the crystal growth studies from melt carried out in space. Here, we shall briefly discuss some of the salient points and try to bring out some correlations between the results

of the various crystal growth efforts from melt that have been published. One has to bear in mind that the details as reported are sketchy at best and some observations that do not conform to the present state of knowledge on crystal growth have not been commented upon or attributed to some unspecified contaminants. Another point to be noted is that there is little evidence of inter-experimental consistencies in the experimental set-up, although this could have been of tremendous value.

2.1. Experimental conditions

For the crystal growth experiments a multipurpose furnace system was used which consists of the furnace, a programmed electronic temperature controller and, in the ASTP experiments, a helium rapid-cooldown system. The basic design of the furnace is shown in figure 2. Each experimenter is provided a set of three stainless steel cartridges. Each cartridge contained the crystal assembly in ampoules to be processed. The cartridges were inserted into three tubular cavities of the furnace and were processed simultaneously. The crystal assemblies were all quite different, to provide for the special thermal environment of the investigator. The heat flow through the crystal to the heat extractor plate was controlled by heat levellers, lateral heat shields and a heat extractor system. Melting was initiated with the application of appropriate power at constant temperature and the crystal melt interface was stabilised by thermal soaking for a predetermined period. After thermal soaking the power system was switched to the cooldown mode. In the ASTP experiments (Gatos *et al* 1977; Boese *et al* 1977) for instance, the cooling rate was 2.4 K/min which was expected to yield a microscopic growth rate ranging from 5 $\mu\text{m}/\text{sec}$ at the beginning to 10 $\mu\text{m}/\text{sec}$ at the end of the controlled cool-

Figure 2. Schematic diagram of the multipurpose furnace used for ASTP crystal growth and solidification experiments (figure 24.1 from Gatos *et al* 1977).

down period. After this period the power was switched off and the furnace temperature decreased gradually to room temperature in the passive mode. In the ASTP experiment helium gas was introduced after roughly 70 min in the passive mode. Room temperature was achieved by this method in 3 hr compared to roughly 20 hr in the *Skylab* missions. The temperature at the hot end of the furnace during the thermal cycling in some of the experiments is shown in figures 3 and 4.

Figure 3. Temperature profile for ASTP experiment MA-060 (figure 21.6 of Boese *et al* 1977).

Figure 4. 'Hot end' temperature cycles for *Skylab* III and *Skylab* IV experiments (figure 3 from Witt *et al* 1975).

2.2. Nature of experiments

The details of the experiments on crystal growth from melt that were carried out in space in the various missions are outlined in table 3. In the ASTP experiment (Gatos *et al* 1977) an elegant method of monitoring crystal growth rate was used by sending current pulses every 4 sec through the growing crystal. This method perhaps had its genesis in an earlier *Skylab* IV experiment (M562) (Witt *et al* 1975) in which it was found that a predetermined mechanical shock given to the growing crystal could be seen in the form of a dopant segregation discontinuity.

We find from table 3 that two types of experiments were carried out in the space mission. Four of these experiments involve growing crystals in container which are called closed tube experiments and two of these involve growing containerless crystals. We shall first analyse the results of the closed tube experiments and then look at the result of the containerless experiments separately.

3. Results

3.1. Closed tube experiments

3.1a. *Morphological observations*: In almost all the closed tube experiments (Gatos *et al* 1977; Witt *et al* 1975; Yue and Voltmer 1975) there was no wetting of the container wall by the melt in the initial stages of crystal growth. This wetting inversion was not predicted. The lack of wetting gave rise to smooth surface features (figures 5-8) and in some cases the crystal came freely out of the containing quartz ampoule. In some cases there was an actual reduction in the crystal diameter in the initial stages of crystal growth reminiscent of 'necking' (figure 8) (Witt *et al* 1975). The later stage of crystal growth was accompanied by bumpy, irregular, morphological features (Gatos *et al* 1977; Witt *et al* 1975). A ridge-like pattern, approximately $0.25\text{ }\mu\text{m}$ in height, was observed in some cases (figures 6-7) which becomes more prominent at the end of the crystal. Attempts to explain the ridge formation on the basis of known phenomena such as wetting and convection induced by surface tension gradients have not been successful (Yue and Voltmer 1975). The low rate of cooling towards the end of the crystal growth experiments could have resulted in smaller temperature gradients, and hence separation of inhomogeneities by constitutional supercooling is possible. The generally accepted interpretation of the irregular features at the end of the crystal growth is attributed to forced confinement of the liquid in the graphite end of the ampoules.

It is to be noted that the temperature of the initial thermal soak period in different missions was different. The melt-solid interface surface was concave to the melt probably because conductive heat transfer is the most important mechanism. It has not yet been properly established what relation the concavity of the interface has to this thermal soaking period. For instance, the necking observed in the Te doped InSb sample in *Skylab* IV (Witt *et al* 1975) in contrast to *Skylab* III was attributed to the higher thermal soaking temperature of the former mission. This could lead to a more concave interface which could lead to

5

6

Figure 5. Germanium crystals regrown in space after removal from the ampoules (a) $\langle 100 \rangle$ direction, (b) $\langle 111 \rangle$ direction (figure 24.3 from Gatos *et al* 1977).

Figure 6. Irregular ridge patterns on the surface of the $\langle 100 \rangle$ Ge crystal regrown in space. (Magnification $\times 123$) (figure 24.4 from Gatos *et al* 1977).

7

8

Figure 7. Part of the Te-doped crystal grown during *Skylab* III (experiment M562) 3.7–5.9 cm from initial regrowth interface. Surface ridges broaden and branch out at late stages of growth (right hand side) (Magnification $\times 6.8$) (from figure 5 of Witt *et al* 1975).

Figure 8. Te-doped crystal grown during *Skylab* IV mission showing ‘necking’ surface ridges appearing on the right hand side (figure 6 from Witt *et al* 1975).

Table 3. Crystal growth experiments in space.

Expt. No.	Mission	Crystals grown					Cooling rate (°C/min)	Soaking conditions	Container	Reference
		Crystal	Dopant (concn. atoms/cm ³)	Orientation	Diameter (mm)	Seed				
M559	Skylab III	Ge	Ga (7.8×10^{19}) Sb (0.42×10^{19}) B (2×10^{19})	$\langle 111 \rangle$	8	Fused grown in $\langle 111 \rangle$ direction	0.6	960° C for 16 hr	Sealed silica tube	Yue and Volmer (1975)
M560	Skylab IV	InSb	Se ($\sim 10^{19}$)	$\langle 110 \rangle$		Czochralski in $\langle 110 \rangle$ direction	0.6		Containerless	Walter (1977)
M562	Skylabs III and IV	InSb	Undoped Te ($\sim 10^{18}$) Sn ($\sim 10^{22}$)	$\langle 111 \rangle$	14	Czochralski in $\langle 111 \rangle$ direction	1.17	80° C for 2 hr (additional after 140 min cooling for 1 hr and a mechanical shock after 90 min of cooling)	Sealed quartz tubes	Witt <i>et al</i> (1975)
M563	Skylabs III and IV	In _{0.9} Ga _{0.1} -Sb	..	..	8	Polycrystalline dendritic ingots	0.6	950° C for 16 hr	Sealed silica tubes	Yee <i>et al</i> (1975)
MA060	ASTP	Ge	Ga, Sn, B (10^{18} - 10^{19})	$\langle 100 \rangle$ and $\langle 111 \rangle$	10, 5	Czochralski grown	2.4	1400 K for 2 hr	Graphite cups at end allowed sending of electrical pulses every 4 sec of 55 milli-second duration of 19.1 A/cm ² current	Gatos <i>et al</i> (1977)

smaller diameter for the growing crystal. However, experiments with $\text{In}_x\text{Ga}_{1-x}\text{Sb}$ showed the opposite results, the ingots obtained in *Skylab* III having a smaller diameter than those in *Skylab* IV. The differences in the latter case were attributed to surface tension effects on the interface concaveness due to increased contamination by impurities at the higher thermal soak temperatures.

3.1b. Direct melt interface observation : Attempts to observe the interface directly in the doped germanium crystals grown in *Skylab* III by etching the crystals in Dash did show a poorly-defined concave interface (to the melt) with the degree of concaveness being less in the space crystal than in the crystal grown on earth under identical conditions. This result agrees with the thermodynamics of a free surface solidification. The low level of doping (table 3) did not allow the interface to be rendered visible by striation etching.

On the other hand etching of the rather heavily doped InSb crystals with a solution containing 1HF (48) and $1\text{CH}_3\text{COOH} + 1\text{KMnO}_4$ (saturated aqueous solution) clearly shows (figure 9) a concave interface. The sharp demarcation line is due to an abrupt decrease in the Te concentration as required for initial regrowth since the distribution coefficient k_0 of Te in InSb is less than 1. Although this result is at first glance a dramatic vindication of the thesis that absence of gravity-induced convection would remove compositional inhomogeneities, one must remember that the heavily-doped crystals of InSb were chosen such that striations could be seen and the original crystal being obtained by the Czochralski method was bound to show rotational striations which would disappear anyway once the crystal was melted.

Gatos *et al* (1977) also carried out a similar experiment in the ASTP series on a Czochralski grown Ge single crystal heavily doped with Ga, and found superficially similar results as shown in figure 10. Three regions can be identified, the original seed position, a central narrow region exhibiting microscopic compositional inhomogeneities and the bottom region exhibiting no compositional fluctuations. The 4-second electrical pulses that were sent can be seen to cause narrow striations in the bottom portion, the gap between these striations converging to zero at the demarcation between the central and the bottom region. This demarcation region therefore actually marks the onset of crystal growth. The central region associated with compositional fluctuation is therefore the region during which thermal soaking took place, the fluctuations being attributed to thermal instabilities of the furnace. These results clearly show that the interfaces marking the origin of the growth interfaces are not so easy to delineate as the crystal growth interfaces are not at the interface where the rotational striations appear prominently. The second thermal soak period introduced in the M562 *Skylab* IV experiment (figure 3) is also associated with a dopant segregation discontinuity.

3.1c. Dopant segregation behaviour : The first gallium-doped germanium crystals grown in *Skylab* III (Yue and Voltmer 1975) were characterised for dopant segregation behaviour by spreading resistance (SR) measurements. Both two-probe and one-probe SR measurements were made as illustrated in figure 11. The calibration for the spreading resistance measurements was obtained from germanium standards. Yue and Voltmer (1975) defined macrosegregation as solute concen-

9

10

Figure 9. Etched cross-section (under dark field illumination) of crystal grown during *Skylab IV* mission (bottom) (expt. M562) in contrast to earth-grown region (top) exhibits no compositional inhomogeneities (Magnification $\times 12$ figure 10 from Witt *et al* 1975).

Figure 10. Etched segment of the $\langle 111 \rangle$ Ge crystal regrown in space (expt. MA-050); note the seed portion (top) and the controlled regrowth portion (bottom) separated by a band of uncontrolled growth and segregation (Magnification $\times 72$) (figure 24.5 from Gatos *et al* 1977).

Figure 11. (a) Top view of the crystal. Dotted lines indicate the path and direction taken by spreading resistance profiles. (b) Side view indicates one point spreading resistance probe with flat depth indicated (figure 4 from Yue and Voltmer 1975).

Figure 12. Comparison of radial resistivity profile of three Ga-doped Ge crystals. The upper curve is for the crystal resolidified in space while the lower curves are for crystals resolidified terrestrially in horizontal (H) and vertical (V) temperature stabilizing positions. All three curves are made with the two-probe spreading resistance methods at μm steps. The radial profiles are measured at 0.5 cm away from original solid-liquid interface (figure 7 from Yue and Voltmer 1975).

tration fluctuation with a periodicity of 100 to 1000 μm while microsegregation had a periodicity less than 100 μm . The solute inhomogeneity was obtained from the resistivity fluctuations using the formula

$$c = 1/[e\mu(\rho)],$$

where c is the solute concentration, e is the electronic charge, ρ is the resistivity of the region whose concentration is to be determined and $\mu(\rho)$ is the resistivity-dependent mobility. In figure 12 the SR measurements of Yue and Voltmer

taken radially (as detailed in figure 11a) are shown for the space grown sample and compared with the SR measurements on two samples grown on earth under the same conditions. These results show that while the ground-grown samples show a macrofluctuation of roughly 15 to 20% the space grown samples show a fluctuation of only 3%.

Microsegregation was quantified by a standard deviation calculation using the formula

$$\sigma_s/\bar{\rho} = \sqrt{\sum_{i=1}^N (\rho_i - \bar{\rho})^2 / N\bar{\rho}^2}$$

where ρ_i s are the resistivity values taken at every $5\text{ }\mu\text{m}$ and $\bar{\rho}$ is the average resistivity in the N sampled regions. From these studies it was observed that while microfluctuation is roughly 2% for the terrestrial samples, it is only 0.4% for space-grown samples.

The longitudinal SR measurements for the space-grown crystal made near the centre and edge of the flat specimen are shown in figure 13. The edge measurements were made at $400\text{ }\mu\text{m}$ from the flat edge. The solid-liquid interface is marked by a sudden rise in resistivity. These results may be compared with similar measurements carried out on a crystal grown horizontally on earth (figure 14). In both the earth and space-grown crystals the axis and edge measurements gave different results showing that perfect unidirectional solidification was not achieved. The reasons for this behaviour were attributed to the non-flatness of the solid-liquid interface and the presence of radial temperature gradients in the ampoules, although gravity-induced convection may be another reason for the terrestrial ampoules. A radial temperature gradient may be another cause and it has not been ascertained whether such a gradient is present or not in the furnace.

The effective segregation coefficient at the solid liquid-interface is given by

$$K_{\text{eff}} = C_s/C_L = \rho_L \mu(\rho_L) / \rho_s \mu(\rho_s),$$

Figure 13. Longitudinal SR resistivity measurements for the space crystal made near the centre and edge of the flat (figure 11 from Yue and Voltmer 1975).

Figure 14. Longitudinal SR resistivity measurements for the ground crystal 4 (CH) (figure 12 from Yue and Voltmer 1975).

where the subscript s denotes solid and L is that region far away from the interface (taken to be the region which did not melt). It was found that $k_{eff} = 0.12$. k_{eff} is expected to be greater than the ratio k_0 obtained at equilibrium which is 0.087 for Ga in Ge (Walter 1977). The remarkable result is that k_{eff} for the space-grown crystal is always larger than the earth-grown crystal. This implies that there is a thicker boundary layer in space all along the interface, and has been attributed to smaller convective mixing in space. It can be seen from figures 14 and 15 that k_{eff} approaches the value of 1.

The ASTP experiments of Gatos *et al* (1977) with a more heavily Ga-doped germanium crystal also shows the same kind of results from the spreading resistance measurements. It was found in addition that after k_{eff} reaches approximately a value of 1 the dopant concentration decreased over a distance of 1 cm instead of maintaining the steady state value. There was again a further increase which was attributed to breakdown of the interface due to constitutional supercooling mentioned earlier which is not a characteristic of growth under zero- g condition but rather is due to the geometry of the growth system. Segregation inhomogeneities was found to be initiated at the facet-off-facet boundary and the onset of the instability occurred significantly earlier in the $\langle 111 \rangle$ direction.

3.1d. Growth rate behaviour : The interface demarcation procedure used in the MA-060 ASTP experiment (Gatos *et al* 1977) enabled quantitative measurements of the growth rate. The growth rate behaviour of various regions of the crystal grown in $\langle 111 \rangle$ direction is given in figure 15. The growth rate behaviour was the same in the space and earth-based experiments showing that conductive heat transfer is the dominant mechanism. The initial transient region in the growth rate which picks up rapidly from approximate $0 \mu\text{m}/\text{sec}$ to approximately $7 \mu\text{m}/\text{sec}$ has led Gatos *et al* (1977) to consider the development of a modified segregation theory.

Figure 15. Microscopic growth rate of the (111) Ge crystal regrown in space (figure 24.8 from Gatos *et al* 1977).

3.2. Alloy solidification

The influence of gravity on alloy solidification (Yee *et al* 1975) using melts of $\text{In}_2\text{Ga}_{1-x}\text{Sb}$ was the purpose of the study of the solidification of these melts in space. The specimens grown in space were all polycrystalline. The freezing was not unidirectional and no compositional profiles were reported. It was found that the number of twin boundaries was dramatically less in the space-processed ingots compared to the earth-processed ones. The number of grain boundaries also appeared to be slightly less in space. There was no significant difference in the void volume or the tendency to form microcracks in the space or earth-based experiments. The voids are concentrated at the initial remelt interface in the earth-based experiments while in the *Skylab* ingots they were more uniformly distributed showing that there is some beneficial effects of gravity which help in eliminating gas bubbles.

3.3. Containerless experiments

Although the lack of melting in some of the closed tube experiments showed that those experiments are essentially containerless we deal in this section specifically with those experiments that were aimed at growing spherical crystals. These experiments were carried out by Walter (1976, 1977).

3.3a. Pulsating growth and non-rotational striations : InSb crystals containing 10^{19} atoms of selenium was prepared on board the *Skylab* IV mission by seeded directional solidification of a pendant drop in approximately (110) direction. High

Figure 16. Etched $\langle 111 \rangle$ A longitudinal section of InSb(Sc) single crystal. Arrow indicates demarcation between Czochralski grown seeds at bottom with rotational striation and space-grown section ; $\langle 111 \rangle$ B solid-liquid facet striations at upper left ; off-facet striations at upper right. Etch pits due to In dislocations (Magnification $\times 17$). Inset (b) melt back interface.

resolution differential etching Walter (1977) of a $\langle 111 \rangle A$ section is shown in figure 16. Three regions to be distinguished are (i) the lower region associated with rotational striations of the Czochralski grown seed, (ii) off-facet striations at right on top and (iii) the striations associated with the formation of a peripheral $\langle 111 \rangle B$ solid-liquid interface facet at left. The off-facet striations extend 5–7 mm into the regrown crystal being very distinct at the beginning of the interface and becoming increasingly shallow further from the interface until it disappears completely. Analysis of the experimental condition led Walter (1977) to rule out mechanical or thermal instabilities or fluid flows including Marangoni flows as the basic mechanism. They proposed a basic underlying mechanism which could be some fundamental property of the growth process. Although the existing theories on growth rate R assume a step function increasing from $R = 0$ to the steady state value R_0 , it was shown earlier in the ASTP experiments (Gatos *et al* 1977) that there is an initial transient region. Due to build-up of a diffusion layer in the initial transient region there may be a decrease of liquidus temperature leading to constitutional supercooling. This will lead to a growth rate which is larger than R_0 leading to an overshoot and reversal in the thermal imbalance. This overstable interaction between solute and temperature fields could lead to oscillatory behaviour. As has already been mentioned similar features observed in the ASTP experiment could be attributed to the above mechanism. Considering that there is no mechanism by which inhomogeneities in density could be evened out in a gravity-free environment, it is perhaps worth looking more closely into the mechanism by which inhomogeneities due to rotational striations which are already present in the sample get evened out in space.

3.3b. Generation and propagation of defects : Czochralski grown single crystal of InSb were partially backmelted so that large drops (15 mm diameter) of unconstrained melt could be directionally solidified by controlled cooldown.

The scanning reflection topographs showed single Bragg reflexes which indicated overall single crystallinity. In-dislocation profiles obtained by etching show a sudden increase in the first few millimeters of space-grown material followed by a steep decrease. There was no particular directionality of dislocation as discerned by Bormann anomalous transmission topography in the earth-grown crystal while those in the space-grown crystals are rectilinear.

Selenium-doped samples grown in $\langle 132 \rangle$ direction had the off-facet striations referred to earlier. A sharp demarcation in the solid melt interface is due to an abrupt change in the selenium concentration ($k_{eff} \sim 0.2$). A scanning reflection topograph shows an overall intensity variation along the axis of the topograph with the highest intensity from the seed region showing greater imperfection in the seed region. Rectilinear dislocations were also observed in these crystals.

The striking difference between the geometry of irregular line defects in seed and rectilinear line defects in space-grown material could be attributed either to the absence of strain field in the more perfect space-grown crystal or to the indication of solidification process that was not disturbed by random convective fluid flow. The termination of the rectilinear dislocations at the surface leads to a continuous decrease of dislocation density along the crystal growth direction further from the solid melt interface.

Acknowledgement

The author would like to thank Professors S Ramaseshan and C N R Rao for the opportunity given especially since it enabled him to learn a lot about crystal growth not only in space but also on earth.

References

- Boese A, McHugh J and Seidensticker R 1977 Appollo Soyuz Test Project, *Summary science report* NASA SP-412 1 313
- Cockayne B 1977 *J. Cryst. Growth* **42** 302 413
- Gatos H C, Witt A F, Lichtensteiger M and Herman C J 1977 Appollo Soyuz Test Project, *Summary science report* NASA SP-412 1 429
- Laudise R A 1974 *J. Cryst. Growth* **24/25** 32
- Walter H V 1976 *J. Electrochem. Soc.* **123** 1098
- Walter H V 1977 *J. Electrochem. Soc.* **124** 250
- Witt A F, Gatos H C, Lichtensteiger M, Lavine and Herman C J 1975 *J. Electrochem. Soc.* **122** 276
- Yee J F, Mu-Ching Lin, Sarma K and Wilcox W R 1975 *J. Cryst. Growth* **30** 185
- Yue J T and Voltmer F W 1975 *J. Cryst. Growth* **29** 329