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Semiconductor Crystal Growth in Crossed Electric and Magnetic Fields

(Center Director's Discretionary Fund Final Report Project No. 93–25)

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INTRODUCTION

The primary objective of this work was to investigate the effects that combined electric and magnetic fields can have on gravitationally driven fluid flow during the bulk growth of selected semiconductor alloys. Although the effects of applying a static magnetic field alone to an electrically conducting melt is well documented,[1 2] the number of studies addressing the effects of combined electric and magnetic fields on solidification is quite limited. Crossed electric and magnetic fields have been applied during the solidification of immiscible binary liquid metal systems.[3] The electromagnetic fields induced a force on the melt opposite to that of gravity. The heavier of the two components, which would normally have settled at the bottom of the container, was suspended during the solidification process. It was found that electromagnetic body forces could lead to a more homogeneous alloy upon solidification. Electromagnetic fields can also be used to induce forced convection in the melt. Forced convection can be used to stir multicomponent melts, enhance the interface stability, and reduce thermal asymmetry at the solidification front.[4]

In the initial part of this study, InGaSb ingots were grown without electromagnetic fields applied. One InGaSb ingot was grown with a large aspect ratio and small diameter. The effect of size reduction on solutal convection in the melt was investigated. A growth cell was then constructed in which crossed electric and magnetic fields could be separately or simultaneously applied. InSb ingots were grown in the cell, and the effects of electromagnetic fields on the ingots were assessed. Experiments were also conducted in which a thermocouple was inserted into the melt and the effects of electromagnetic fields were examined in situ.

InGaSb CRYSTAL GROWTH

Two $In_{0.2}Ga_{0.8}Sb$ crystals were grown in the absence of applied electric and magnetic fields. The starting materials were prepared by loading the appropriate stoichiometric amounts of 6N grade indium, gallium, and antimony into fused silica tubing. The materials were homogenized in a rocking furnace before being loaded into fused silica growth ampoules. The ampoules were placed inside vertical directional solidification furnaces for which the desired thermal profiles had been previously determined. Crystal growth occurred when the growth ampoules were translated with respect to the furnaces.

The first ingot was 12.5-cm long and 0.9 cm in diameter and was grown at a rate of 0.12 μm/s. After growth, the ingot was removed from the ampoule and was sandblasted to reveal the grain structure. The ingot was observed to be polycrystalline, with several grains and twins observed. This result is consistent with previous attempts to grow $In_xGa_{1-x}Sb$ with $x \geq 0.1$.[5] The second $In_{0.2}Ga_{0.8}Sb$ ingot grown was 23 cm in length and 0.5 cm in diameter and was grown at a rate of 0.10 μm/s. The small diameter was used to reduce convection in the melt as much as practically possible by reducing this dimension of the system. The temperature profile of the furnace was determined by a thermal probe prior to crystal growth and is shown in figure 1. The melting temperatures of the endpoint binary compounds (GaSb and InSb) are also shown. The temperature of the solid-liquid interface can vary between these temperatures, as the composition of $In_xGa_{1-x}Sb$ can vary continuously during growth. After growth, the axial composition of the ingot was measured by x-ray energy dispersion spectrometry (EDS). The results are shown in figure 2. The InSb mole fraction changed continuously during growth. The axial profile, with the exception of the anomaly observed at approximately 16 cm, is indicative of complete mixing in the melt during growth.

Even with a reduced ampoule diameter, gravity-induced buoyancy-driven convection dominates the solute transport in the melt. The sudden increase in InSb mole fraction at about 16 cm is most likely the result of constitutional supercooling. Constitutional supercooling can occur when

$$\frac{G}{V} \geq \frac{C_s m}{D} \frac{(k_i - 1)}{k_i} ,$$

where G is the equilibrium temperature gradient in the melt at the interface, V is the growth velocity, C_s is the solute concentration in the solid, m is the slope of the liquidus at the interfacial composition as determined by the phase diagram, k_i is the interfacial segregation coefficient, and D is the diffusion coefficient of the solid in the liquid. Calculations[6] indicate that at the growth rate of 0.10 µm/s, a temperature gradient of approximately 35 °C/cm was needed to avoid constitutional supercooling for an InSb melt concentration of 0.5. Although the thermal probe data obtained prior to the experiment measured a temperature gradient larger than 35 °C/cm, the sample has a higher thermal conductivity than the thermal probe and changes the heat transport conditions. Thus, it is quite possible that the criterion for constitutional supercooling was met. According to the InGaSb phase diagram,[7] an InSb concentration of 0.5 in the melt is in equilibrium with an approximately 0.1 InSb concentration in the solid. Thus, a sudden freezing of the melt ahead of the solid-liquid interface is consistent with the data depicted in figure 2.

APPLICATION OF ELECTRIC AND MAGNETIC FIELDS

Cell Design and Construction

A cell was designed in which electric currents and magnetic fields could be applied separately or in combination to a semiconductor melt. A schematic diagram of the cell is shown in figure 3. The entire cell is enclosed in a rectangular quartz ampoule that is open on the top. Molybdenum rods were placed in rectangular graphite posts on the sides of the cell. Molybdenum rods were chosen as electric conductors because they could withstand the high temperatures to which the cell was subjected and because they had considerably higher electrical conductivity than either the graphite or the semiconductor melt. This was necessary to insure that the electric current was uniform across the melt and independent of the melt height. The graphite provided a high-temperature electrically conducting material at the ends of the cell and electrically insulating quartz on the sides of the cell made sure that the current traveled through the melt. Boron nitride was used at the bottom of the cell and was shaped to enhance self-nucleation during crystal growth. The outer dimension of the cell were 1.3 by 4.2 by 14 cm, and could accommodate melt dimensions of 0.9 by 2.0 cm and up to 12 cm in height. The cell was placed inside a cylindrical quartz tube that was placed inside a transverse electromagnet that could provide magnetic fields up to 0.5 Tesla. The directions of the applied electric and magnetic fields are shown at the top of figure 3. Electric and magnetic fields could be used in combination to produce a Lorentz force in either the upward or downward direction, either opposing or adding to the force of gravity.

Results

Several tellurium-doped InSb crystals were grown in the cell described above. InSb was chosen for initial crystal growth experiments because it has a lower melting temperature than InGaSb, can be grown at a faster growth rate, and does not suffer from complicating solutal effects. Several InSb crystals were grown without electric or magnetic fields applied. A hydrogen/helium gas mixture was made to flow through the growth system in order to reduce oxidation of the InSb. The grown InSb ingots were shiny in appearance and did not stick to the quartz, graphite, or boron nitride components of the growth cell. However, the ingots were polycrystalline with several large grains observed. Although the growth cell allows for the simultaneous application of perpendicular electric and magnetic fields, it does not provide the optimum thermal profile to the growing crystals. The thermal profile is asymmetric, in part because the quartz and graphite have significantly different thermal properties. The inherent asymmetry in the cell may have increased the possibility of nucleation at the cell walls and contributed to the polycrystalline nature of the material.

One InSb ingot was grown while electric current pulses of 30 A in magnitude and 1 min in duration were periodically applied. The purpose of the experiment was to assess the effect, if any, of passing current parallel to the growth interface. This is in contrast to the standard interface demarcation procedure, where electric current travels perpendicular to the growth interface, from the solid to the liquid phase (or vice versa). After growth, the appropriate etch was applied,[8] but no effect of the applied current was optically detected. Another crystal growth experiment consisted of applying no magnetic or electric fields during the first half of growth and then applying magnetic and electric fields simultaneously for the second half. The current and magnetic field were applied such that the resulting Lorentz force was pointing upward, in the opposite direction as gravity. Although no well-defined demarcation line was observed when the electromagnetic force was turned on, the corners of the ingot had pulled away from the cell enclosure during the second half of growth. The ingot was polycrystalline, with several large grains and twins. An attempt was also made to grow an $In_{0.2}Ga_{0.8}Sb$ crystal in combined electric and magnetic fields. However, it was not possible to maintain a constant current density during the 16 days required to grow the crystal. Also, the magnet shut itself off unexpectedly during growth. It was not possible to correlate the electromagnetic field changes to observations of the grown ingot.

A series of measurements was made with a thermocouple directly inserted into an InSb melt. The purpose was to assess the temperature response of the melt to applied electric and magnetic fields. The outer diameter of the thermocouple sheath was 20/1000 of an inch and the end of the thermocouple was coated with aluminum oxide to prevent it from reacting with the InSb melt. The first experiment measured the effectiveness of a static magnetic field in suppressing time-dependent convection. A destabilizing temperature gradient (hotter on the bottom than on the top) of approximately 3 °C/cm was applied. For magnetic field strengths up to 0.04 Tesla, temperature fluctuations were observed. Above 0.04 Tesla, the thermocouple gave a time-independent response. Of course, in general the magnetic field strength required to suppress time-dependent convection is a function of the thermal profile and geometry of the system.

In another set of experiments, a 0.4-Tesla magnetic field was applied and 4 A of electric current was periodically turned on and off. The temperature transients measured near the bottom of the cell are shown in figure 4. When the current was applied, the temperature increased sharply and, when the current was shut off, the temperature changed back to its previous value. The temperature changes can be explained as a result of fluid flow in the cell. The cell is in a stabilizing temperature gradient, with the temperature hotter on the top than on the bottom. When the current is applied, electromagnetic forces cause warmer fluid to flow from the top to the bottom of the cell. When the thermocouple was placed at the top of the cell, the application of current caused the temperature to decrease because cooler fluid was flowing up from below. Beyond the initial transient, the application of 0.4 Tesla and 4 A did not cause time-dependent convection in the melt. However, sinusoidal temperature oscillations were observed in the middle of the melt when the current was increased to 6 A. Further increases in the current caused the time-dependent temperature fluctuations to become even more pronounced.

SUMMARY

This project has demonstrated that crossed electric and magnetic fields can generate a number of interesting effects during semiconductor crystal growth. A static magnetic field was shown to reduce time-dependent convection, in agreement with previous results.[9] It was found that obtaining a uniform and steady electromagnetic force in opposition to the force of gravity, in attempt to reduce convection, was difficult to achieve. Any inhomogeneity in the applied electric or magnetic fields or in the thermal conditions can lead to forced convection in the melt, ahead of the solid-liquid interface. However, controlled forced convection can itself be very beneficial to the crystal growth process in a number of circumstances. The results of this project led to a proposal entitled "Electromagnetic Field Effects in Semiconductor Crystal Growth," which was accepted for funding by NASA's Microgravity Science and Application Division.

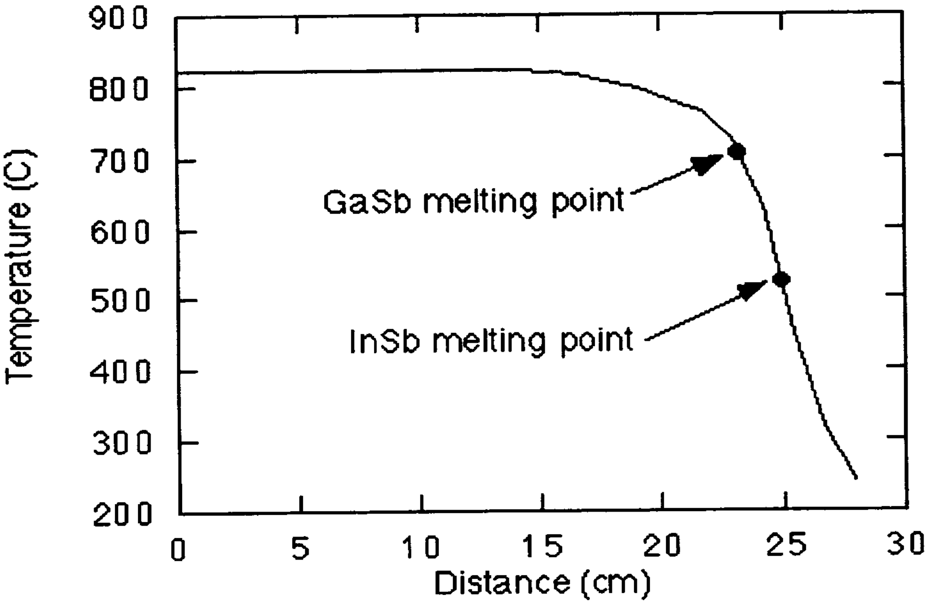


Figure 1. Furnace temperature profile used for the InGaSb crystal growth experiment.

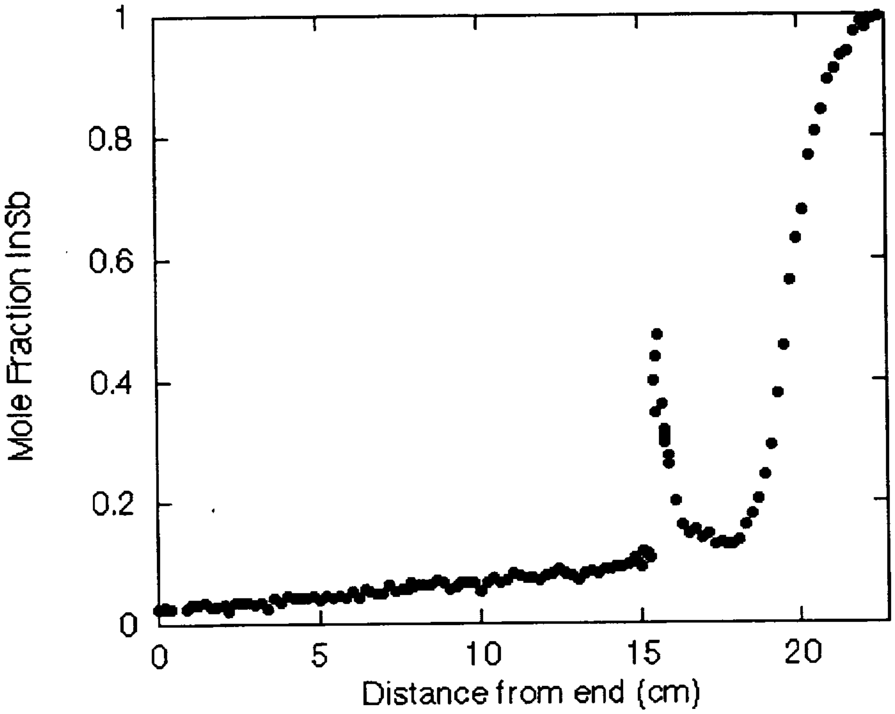


Figure 2. Axial composition profile of 23-cm long by 0.5-cm diameter InGaSb crystal. The sudden increase in InSb mole fraction at about 16 cm may indicate constitutional supercooling and a breakdown of the solid-liquid interface.

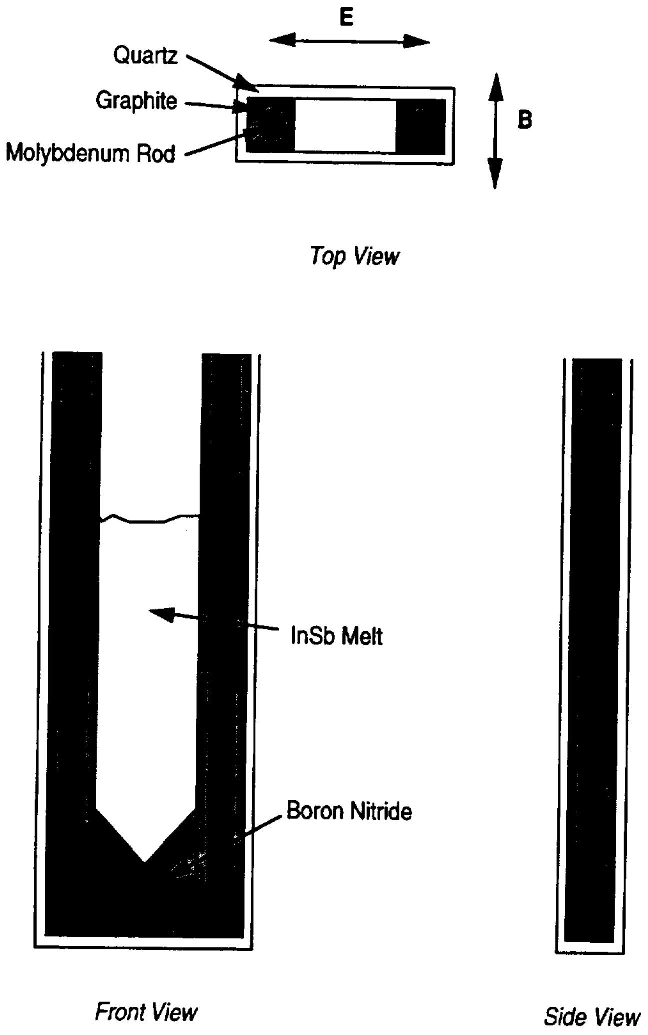
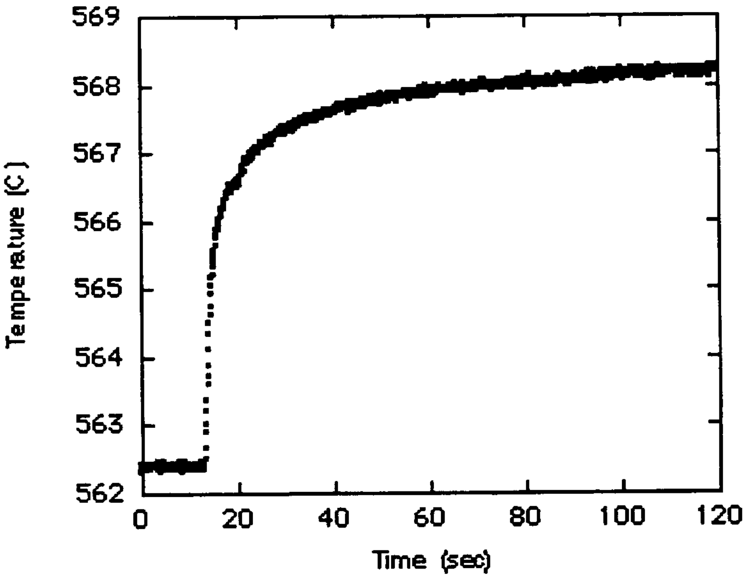
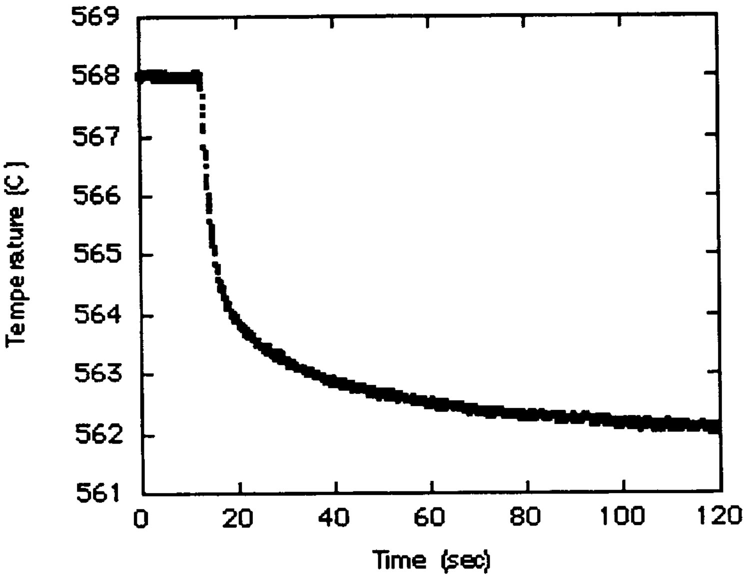


Figure 3. Schematic diagram of the growth cell. The electric current travels down the molybdenum rod and through the melt. The magnetic field is perpendicular to the current direction.



(a)



(b)

Figure 4. (a) Temperature transient when 4 A of current are applied to the cell in a 0.4-Tesla magnetic field. (b) Temperature transient when the current is shut off.

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APPROVAL

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The information in this report has been reviewed for technical content. Review of any information concerning Department of Defense or nuclear energy activities or programs has been made by the MSFC Security Classification Officer. This report, in its entirety, has been determined to be unclassified.

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Director, Space Science Laboratory

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A unique growth cell was designed in which crossed electric and magnetic fields could be separately or simultaneously applied during semiconductor crystal growth. A thermocouple was inserted into an InSb melt inside the growth cell to examine the temperature response of the fluid to applied electromagnetic fields. A static magnetic field suppressed time-dependent convection when a destabilizing thermal field was applied. The simultaneous application of electric and magnetic fields resulted in forced convection in the melt. The InSb ingots grown in the cell were polycrystalline. An InGaSb crystal, 0.5 cm in diameter and 23-cm long, was grown without electromagnetic fields applied. The axial composition results indicated that complete mixing in the melt occurred for this large aspect ratio.

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