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Eddy Current Sensing of Vertical Bridgman Semiconductor Crystal Growth

A Dissertation

Presented to

the Faculty of the School of Engineering and Applied Science

University of Virginia

In Partial Fulfillment

of the requirements for the Degree

Doctor of Philosophy

Mechanical and Aerospace Engineering

by

Kumar Pradeepa Dharmasena

May 1996
APPROVAL SHEET

The dissertation is submitted in partial fulfillment of

the requirements for the degree of

Doctor of Philosophy in Mechanical and Aerospace Engineering

Kumar P. Dharmasena, Author

This dissertation has been read and approved by the examining committee:

H.N.G. Wadley, Dissertation Advisor
D.M. Elzey
W.A. Jesser
L.G. Richards
E.A. Thornton, Chairman

Accepted for the School of Engineering and Applied Science:

R.W. Miksad
Dean, School of Engineering and Applied Science

May 1996
Abstract

The vertical Bridgman process is a directional solidification technique commonly used to grow large diameter semiconductor crystals. Low thermal conductivity semiconductors grown by this technique (e.g. Cd$_{1-x}$Zn$_x$Te alloys) are consistently poor in quality with low yields and high production costs. This has been attributed to an inadequate understanding of the growth process and the inability to measure and control the melt stoichiometry, solidification velocity and the shape of the liquid-solid interface. It has stimulated a search for non-invasive sensors able to function in the high temperature crystal growth environment and provide measurements of the melt stoichiometry, the growth velocity and the interfacial shape. The significant difference in the electrical conductivity of solid and liquid semiconductors at their melting point has led to an interest in the use of eddy current sensors for these applications.

Electromagnetic finite element methods (FEM) were used to analyze radio frequency electromagnetic field interactions with liquid-solid semiconductor interfaces. From this, the multifrequency response of two encircling eddy current sensor designs to the passage of a liquid-solid interface was obtained. The liquid-solid interface shape, the frequency of excitation and the liquid/solid electrical conductivities were then varied to identify the factors controlling sensor performance. Two approaches for discriminating the interface position from its shape were identified. The first was based on a comparison of the low and high frequency impedance responses. The second used the observation of an inflection point in the sensor response as an interface passed through the sensor. Its intermediate frequency dependence was a strong function of interface shape while the high frequency asymptotic response depended only on interface location. A model system was used to experimentally validate the proposed sensor concepts. Excellent confirmation of the FEM results was obtained up to a frequency of 3MHz. Above this, unmodelled test circuit effects began to influence the sensor measurements. A non-invasive, high temperature sensor was designed, fabricated and installed in a commercial 6-zone vertical Bridgman furnace and used to monitor several Cd$_{0.96}$Zn$_{0.04}$Te growth runs. Analysis of
the eddy current sensor data prior to growth indicated the melt to be non-stoichiometric, consistent with the evaporation of Cd. Analysis of the sensor’s response during growth revealed a highly convex interface shape that propagated at a significantly slower velocity than expected. These observations were the result of solidification occurring in an unintended (cooler) region of the thermal gradient due to the lower melting temperature of the non-stoichiometric melt. Eddy current sensors are found to be well suited for characterizing solidification processes and clearly help to obtain a better understanding of the growth process. With minor modifications, they could also be used for on-line monitoring and feedback control of directional solidification.
Acknowledgments

I would like to express my thanks to my family, friends and colleagues who stuck with me through good times and bad till I finally saw the light at the end of the tunnel. First and foremost, my advisor Prof. Haydn Wadley whose sound advice, encouragement and patience were immeasurable. I feel indeed a privilege for having got the opportunity to work under his guidance. I also wish to thank Profs. Earl Thornton, Larry Richards, Bill Jesser and Dana Elzey for having served on my advisory committee and provided constructive criticism of my dissertation.

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<th>Symbol</th>
<th>Description</th>
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<tr>
<td>$A$</td>
<td>Magnetic vector potential</td>
</tr>
<tr>
<td>$B$</td>
<td>Magnetic flux density</td>
</tr>
<tr>
<td>$D$</td>
<td>Displacement field, ampoule diameter</td>
</tr>
<tr>
<td>$E$</td>
<td>Electric field intensity</td>
</tr>
<tr>
<td>$F$</td>
<td>Energy functional</td>
</tr>
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<td>$H$</td>
<td>Magnetic field intensity</td>
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<tr>
<td>$I$</td>
<td>Current</td>
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<tr>
<td>$J$</td>
<td>Current density</td>
</tr>
<tr>
<td>$M$</td>
<td>Global finite element matrix</td>
</tr>
<tr>
<td>$N$</td>
<td>Number of coil turns</td>
</tr>
<tr>
<td>$R$</td>
<td>Ampoule radius, reflection coefficient</td>
</tr>
<tr>
<td>$V$</td>
<td>Voltage</td>
</tr>
<tr>
<td>$Z$</td>
<td>Acoustical impedance, Electrical impedance</td>
</tr>
<tr>
<td>$f$</td>
<td>Frequency (in hertz)</td>
</tr>
<tr>
<td>$g$</td>
<td>Gain</td>
</tr>
<tr>
<td>$h$</td>
<td>Heat transfer coefficient, position of interface</td>
</tr>
<tr>
<td>$k$</td>
<td>Thermal conductivity</td>
</tr>
<tr>
<td>$r$</td>
<td>Secondary (pick up) coil radius</td>
</tr>
<tr>
<td>$t$</td>
<td>Time</td>
</tr>
<tr>
<td>$z$</td>
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<td>$\delta$</td>
<td>skin depth</td>
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<td>Dielectric constant or permittivity</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>Electrical conductivity</td>
</tr>
<tr>
<td>$\theta$</td>
<td>Interface shape convexity parameter</td>
</tr>
<tr>
<td>$\mu$</td>
<td>Magnetic permeability</td>
</tr>
<tr>
<td>$\phi$</td>
<td>Phase</td>
</tr>
<tr>
<td>$\omega$</td>
<td>Frequency (in rad/s)</td>
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### Suffixes

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<thead>
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<th>Symbol</th>
<th>Description</th>
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<tr>
<td>$l$</td>
<td>liquid</td>
</tr>
<tr>
<td>$p$</td>
<td>primary</td>
</tr>
<tr>
<td>$s$</td>
<td>secondary, solid</td>
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Chapter 1: Introduction

Compound single crystals of CdTe and Cd$_{1-x}$Zn$_x$Te alloys are used as infrared transparent substrates for the epitaxial deposition of Hg$_{1-y}$Cd$_y$Te infrared detector films used in the high performance focal plane arrays of second generation infrared imaging systems[1-7]. The single crystal substrates needed for infrared detector applications are normally "mined" from large polycrystalline boules obtained by the directional solidification of Cd$_{1-x}$Zn$_x$Te melts, predominantly using a vertical or horizontal Bridgman process[8-21]. In spite of extended efforts by many groups to improve the Bridgman crystal growth process (i.e. by experimenting with furnace temperature profiles, ampoule geometries/materials, charge material purity/stoichiometry and furnace translation rates), the ultimate yield of Cd$_{1-x}$Zn$_x$Te suitable for large area substrates (e.g. 4cm x 6cm) needed today has remained very low (<10%)[22-26].

The reasons for yield loss are many and complex, but among the most important are the polycrystalline nature of the as-grown boules (Figure 1.1), macrosegregation of Zn in Cd$_{1-x}$Zn$_x$Te alloys [27-31], high dislocation densities (>10$^5$cm$^{-2}$)[32-37], and low (<0.65 of theoretical) infrared transmission due to inhomogeneities (principally Te precipitates and/or inclusions greater than 10μm in diameter)[38,39]. The poor yield directly affects the affordability of these substrates; it also results in the need for extensive characterization and screening which further increases the cost of substrate manufacture[40,41]. Since most of the yield loss is associated with "defects" that form during the solidification process, intensive efforts are under way to improve growth technology [42-53].

To a greater, or lesser extent, many other semiconductor materials have less than theoretical yield and suffer from variable quality. Single crystal slices or wafers which are used as substrates for fabricating electronic/optoelectronic devices and integrated circuits are obtained from these bulk semiconductor crystals. Their performance also often depends sensitively on the purity, perfection and homogeneity of the bulk crystals. In fact, semiconductor devices are almost always fabricated on single crystal substrates because
grain boundaries adversely influence the transport of charge carriers and, therefore, device performance\cite{11}. The electronic properties of semiconductors are also intimately related to the degree of crystalline perfection which can be realized in practice. As a result, crystal growth technology has witnessed an ever increasing demand for higher yields and better quality at lower cost. Increasing the wafer/substrate size by growing larger boules has become an important cost reduction strategy. However, these larger diameter samples can be even more difficult to grow. For example, to minimize dislocation densities during crystal growth requires the existence of low radial temperature gradients. As the diameter increases, it becomes more difficult to remove heat through the solid, the gradients increase and it becomes more difficult to obtain high quality material. Since manufacturing costs decrease dramatically with increasing wafer size, the recent trend to increase the diameter of single crystals increases the likelihood of introducing defects and reducing quality.

![Diagram of a Cd$_{0.96}$Zn$_{0.04}$Te ingot grown by the vertical Bridgman process.](image)

Figure 1.1 A photograph of a typical Cd$_{0.96}$Zn$_{0.04}$Te ingot grown by the vertical Bridgman process.
The growth of large single crystals with high compositional uniformity, low concentrations of stoichiometric/point defects, and high crystalline perfection also becomes increasingly difficult as one progresses from elemental semiconductors such as Si and Ge to III-V (e.g. GaAs) and II-VI (e.g. CdTe) compound semiconductors because of a decrease in their thermal conductivities [54,55]. For instance, dislocation-free Si crystals can now be routinely grown at twelve-inch diameter, GaAs crystals up to four inches in diameter (but containing high dislocation densities) have been manufactured, but CdTe has only been solidified as large-grain polycrystalline three-inch diameter ingots.

A measure of the degree of difficulty encountered in the growth of elemental and compound semiconductors can be assessed by comparing the thermal conductivities, $k$, the critical resolved shear stresses, CRSS (extrapolated to near the melting point), and the stacking fault energies, SFE, of selected semiconductors, Table 1.1.

Table 1.1: Thermophysical and mechanical properties of selected semiconductor materials adapted from [11]

<table>
<thead>
<tr>
<th>Element or Compound</th>
<th>Melting point (°C)</th>
<th>$k$ (W/cmK)</th>
<th>CRSS(MPa)</th>
<th>SFE(ergs/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si</td>
<td>1420</td>
<td>0.21</td>
<td>1.85</td>
<td>70</td>
</tr>
<tr>
<td>Ge</td>
<td>960</td>
<td>0.17</td>
<td>0.70</td>
<td>63</td>
</tr>
<tr>
<td>GaAs</td>
<td>1238</td>
<td>0.07</td>
<td>0.40</td>
<td>48</td>
</tr>
<tr>
<td>CdTe</td>
<td>1092</td>
<td>0.01</td>
<td>0.11</td>
<td>10</td>
</tr>
</tbody>
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The fundamental properties identified in Table 1.1 (melting point, thermal conductivity, critical resolved shear stress, and stacking fault energy) provide guidelines to determine which growth technique is the most suitable for a particular semiconductor, whether large single crystals can indeed be grown, and whether the crystals are likely to be dislocation-free at the diameter required. Materials with low thermal conductivities are especially difficult to grow because of the large temperature gradients that arise during processing. These gradients result in thermal expansion stresses. Dislocations are then generated in the material when it is stressed beyond the critical resolved shear stress.
(CRSS). Thus, materials with low thermal conductivities are more prone to stress development and those which have low CRSS values provide a reduced resistance to dislocation formation. Smaller stacking fault energies imply a smaller energy penalty for twin boundary formation and result in a tendency for twin formation during growth. Of the four materials shown in Table 1.1, CdTe has the lowest thermal conductivity, the smallest CRSS, and the least stacking fault energy and is understandably the hardest to grow. The problem is further compounded by the high volatility of one of its components (Cd) at the melting temperature.

Since the majority of undesirable defects are incorporated into the crystal during the growth process, a more "scientific" understanding of the crystal growth process has been needed by the crystal growth community. With the growing availability of high speed computers, one approach has sought to model and simulate the crystal growth process. Several groups have developed one and two dimensional models for the solidification process[10,14,30,42-46,52-53,56-95]. A complete model requires incorporating heat transport by conduction, convection and radiation, analyzing convective flows within the melt, as well as addressing the moving (liquid-solid) boundary and the segregation of a dilute dopant, all as a function of growth conditions (i.e. the incident heat flux distribution and crystal pull (or furnace translation) rate). The solid-interface shape is thought to be a key factor in determining the quality of crystals grown by directional solidification. Other modelling work has sought to calculate the stresses in the crystals and predict the dislocation density and its distribution[96-105]. Significant progress has been made in each of these areas and the ability now exists to simulate a (virtual) growth process with prescribed heat flux distributions (in space and time) applied to the ampoule wall boundary. However, there are very limited experimental results available to validate these models.

A second parallel approach (the thrust of this work) has been to develop sensor technologies for in-situ monitoring of the crystal growth process. The most critical parameters of the growth process to measure include, the melt's composition and
temperature, the degree of convection in the melt, the nucleation of solid on cooling, the liquid-solid interface velocity (i.e. the interface's position within the ampoule as a function of time), the interface's curvature (which affects the probability of secondary grain nucleation and successful competitive growth) and the temperature within the cooling solid. The challenge has been to design sensors which can operate at high temperatures and do not perturb the growth conditions while being able to measure the thermal field, characterize the solid-liquid interface, examine the convective flows within the melt etc. These sensors must therefore rely on differences in the properties of the crystal and melt that can be non-invasively probed. When developed, they promise to provide deeper insights into the growth process, aid model development and may become the basis of new technologies for on-line sensing and, perhaps, feedback control of the growth process[106,107].

In this thesis, non-contact, non-invasive eddy current sensors to characterize the melt and monitor the solid-liquid interface (position and shape) during vertical Bridgman crystal growth of semiconductors will be designed and tested. The sensor concept relies on the large differences in the electrical conductivities of the solid and liquid phases of most semiconductor materials[108], and the non-invasive detection of these using electromagnetic induction concepts.

The thesis is organized as follows: Chapter 2 presents a brief overview of semiconductor crystal growth methods and a synopsis of modelling work. In Chapter 3, the feasibility of various sensor techniques for detecting solid-liquid interfaces are reviewed and assessed. Chapter 4 outlines the fundamental theory of eddy current sensing and the formulation of an electromagnetic finite element modelling approach based on Maxwell's equations to aid in the sensor design. The results of electromagnetic finite element analyses for two selected sensor designs are presented in Chapter 5 for a range of elemental and compound semiconductors. Room temperature experimental results with a bench-top test system are presented in Chapter 6 and used to check the validity of the finite element results. Analysis protocols for discriminating the interface position effect
from the interface shape effect are developed in Chapter 7. Chapter 8 first describes the
design, construction and installation of a high temperature eddy current sensor within a
multi-zone vertical Bridgman crystal grower, then gives the details of the monitored
growth experiments and finally presents the characterization of 72mm diameter
Cd$_{0.96}$Zn$_{0.04}$Te ingots grown in the furnace. Chapter 9 presents the discussion of the
results followed by conclusions in Chapter 10.
Chapter 2: Melt Crystal Growth

2.1 Introduction

Bulk semiconductor single crystals are predominantly obtained by growth from a melt[109-113]. The physical and chemical properties of these melts (such as melting temperature, evaporation rate, and chemical reactivity), to a great extent determine the conditions under which single crystal growth is possible. The coupling of heat and mass transfer with melt flow strongly influences the quality of crystals grown from the melt under these conditions.

It is important to recognize that crystal growth phenomena occur on at least two quite different length scales; at the continuum scale, equilibrium between the crystal and the surrounding medium is governed by transport properties at a macroscopic level (diffusion models in the crystal and Navier-Stokes equations in the melt) while the kinetic phenomena associated with growth of the interface occur on an atomic scale. A crystal growth process is usually analyzed at the continuum level first. This then establishes the local environment where the kinetic processes are responsible for twin/grain boundary formation and solute entrainment. After solidification, the continuum thermal field is responsible for the development of stress which establishes the conditions that favor formation of line defects (dislocations).

2.2 Melt growth methods

Three melt growth techniques are most commonly used for semiconductor bulk crystal manufacture: a) the Czochralski method, b) the floating-zone technique, and c) the Bridgman method. They differ in the ways in which the melt and the growing crystal are supported. Both the Czochralski and the floating zone methods are meniscus defined systems since the crystal is not in contact with a growth crucible/ampoule. In contrast, the vertical Bridgman technique is a confined growth system where the crystal grows within a crucible/ampoule.
2.2.1. The Czochralski method

The Czochralski method is the most commonly used technique for the fabrication of elemental semiconductor bulk crystals, Figure 2.1[114]. The melt is contained in a crucible but the solid-liquid interface and the growing crystal are not in contact with the container walls. The charge can be melted by resistance or induction heating. After melting the starting materials, a small single crystal seed is dipped in the free surface of the melt and slowly withdrawn. The seed is kept slightly cooler than the melt, thereby allowing material to solidify on the seed as it is withdrawn, thus forming a large single crystal. Both the crystal and the crucible are generally rotated in order to minimize thermal asymmetry.

Figure 2.1 The Czochralski silicon crystal-growth technique with rf heating, crystal pull and crystal rotation (from [114]).
In this process, both the diameter of the growing crystal and the solid-liquid interface have to be controlled. (Details of a study which explored eddy current sensing concepts to monitor both of these key parameters are given in Ref. [115].)

The Czochralski process may be readily used for pulling crystals of semiconductors which do not have a high melting temperature and/or which melt congruently. Of the III-V compounds, indium, gallium and aluminum antimonides belong to this group. Indium and gallium arsenides and phosphides however have a relatively high vapor pressure at their melting point. To handle the volatile constituents of these compound semiconductors, an oxide material (usually B₂O₃) is often layered over the melt in a variant of the Czochralski process referred to as the liquid encapsulated Czochralski (LEC) growth technique.

A drawback of the Czochralski process is the contamination of the high purity melt as a result of dissolution of the fused silica crucible. In the case of silicon growth, this leads to crystals containing high interstitial oxygen and electrically active boron impurity concentrations. Crucible-free, floating-zone growth overcomes this deficiency and is a highly developed commercial technique today.

2.2.2. The floating-zone technique

In the floating zone system (Figure 2.2), no crucible is used. A molten pool is formed by a circumferential heat source that separates a melting polycrystalline seed rod and a solidifying cylindrical crystal. In small-scale, resistively heated zones, the melt pool is held in shape by capillary forces and hydrodynamic stresses caused by flow in the melt[114].

Large diameter industrial floating zone systems have been developed using radio-frequency induction heating elements shaped so that the induction coil has a smaller diameter than the growing crystal[114]. These systems are used for growth of high purity semiconductor materials, such as high resistivity silicon and germanium.
2.2.3. The vertical Bridgman method

The Bridgman method is often used for semiconductors that have high vapor pressures (e.g. II-VI compounds such as CdTe) where the crystals can only be grown in closed ampoules (Figure 2.3). In this confined growth method, the charge material is loaded into a crucible/ampoule, melted and re-solidified by varying the temperature field either by translating the ampoule through the furnace or moving the furnace relative to a stationary ampoule. The vertical gradient freeze technique is a variation of the vertical Bridgman method where instead of causing a relative movement between the sample and the furnace, the temperature of the furnace is changed by a time-dependent variation of the heater power. The Bridgman method is not used for crystal growth of materials that significantly expand on solidification. Unlike the Czochralski growth process, the solidification process cannot be “optically” observed during Bridgman growth and has led to a lack of feedback control of the growth process. Trial and error methods have been used as a practiced “art” in the design process to select optimum furnace temperature profiles and translation rates for each material system.
Figure 2.3 The vertical Bridgman furnace, its temperature profile, and the charge contained in a sealed ampoule (from [116]).

2.3 Crystal Growth Modelling

The primary aim of modelling a crystal growth process is to provide a more scientific understanding of the different factors which may influence the quality of the crystal in order to design a better growth process. An ideal model should be able to,

a) Predict the shape of the growing crystal.

b) Relate the quality of the crystal lattice to the stress generated (in the crystal) during growth.

c) Account for melt flow and the influence of the flow patterns on the crystal composition.

d) Relate the factors in a), b) and c) to the control variables of the process such as the temperature profile and furnace movement rates.

The physical phenomena which play a significant role in affecting the crystal quality during bulk growth are illustrated in Figure 2.4. The temperature distribution in a crystal determined by the heat flux is a complicated function of the heat exchange with its
Figure 2.4 Schematic representation of the physical phenomena related to bulk growth that affect crystal quality [117].
environment. Analysis of the heat transfer which includes conduction in all phases, convection in the melt, latent heat generation at the interface, and conduction, convection and radiation between various parts of the growth system geometry enables the calculation of the thermal field. Figure 2.5 shows these basic mechanisms of heat exchange in a typical liquid-encapsulated Czochralski growth arrangement of the thermal field.

Figure 2.5 Heat transfer mechanisms during Czochralski crystal growth (from [118]).
A detailed analysis of the melt will enable the segregation behavior to be investigated and its effect on the chemical composition (dopants, impurities etc.). A thermal history of the crystal would allow the determination of the stresses in the crystal using either thermoelastic or viscoplastic models. It would then allow the quality of the crystal to be evaluated in terms of the dislocations, residual stresses and defects.

Several types of thermal analyses of Bridgman type crystal growth systems have been developed. They include one-dimensional[64,66-68,70-71,83-84], two-dimensional [10,14,30,42-44,53,56,64-69,71-76,83,86] and more recently, three-dimensional[45,46] models. Some formulations used dimensional variables with properties specific to a chosen material system[67]. Others used non-dimensional representations which could be applied to any material system[64-66,70-72]. Solution techniques range from analytical methods[64,68-71,83-84,86] (with many simplifying assumptions) to numerical (finite difference[65-67], finite element[10,14,30,42-46,52-53,73-75,81], and boundary element[82]) approaches that more realistically capture the real geometry as well as the essential heat/mass transport and thermodynamically driven processes.

The principal results obtained from a 1-D analysis are the axial temperature gradient at the growth interface and the axial position of the growth interface. The differences among the various 1-dimensional analyses pertain to specific effects incorporated into the various thermal models such as the inclusion of the crucible[67,83-84], generation of latent heat at the growth interface[64,67,70-71,83-84], unequal melt and crystal thermal conductivities[67,70-71,83-84], addition of a thermal gradient control section[66-67,70-71], and translation rate of the ampoule[64,66-67,70-71,83-84]. Convection was considered to be a non-dominant mode of heat transfer with insignificant influences on the thermal field.

In order to obtain the interface shape, two-dimensional models are required. Two dimensional models allow the radial temperature variations to be predicted in addition to axial temperature variations. Chang and Wilcox[64] pioneered the use of analytical methods to investigate the effects of changes in operating process parameters and material
parameters on the interface position and shape. A two dimensional cylindrical model was analyzed assuming constant properties, an infinitely long cylindrical sample (with no ampoule), a heater section and a cooler section (both at constant temperatures), and a constant heat transfer coefficient between the sample and its surroundings. Convection in the melt and latent heat effects were neglected. They predicted (with solutions obtained in Fourier-Bessel infinite series form) that the interface shape was very sensitive to its position within the furnace, and its position was determined by the heater and cooler temperatures. Lowering the heater and/or cooler temperatures moved the interface upwards into the heater section producing a convex shape. Increasing the heater and/or cooler temperatures had the opposite effect, i.e. a concave interface occurring in the cooler section. The sensitivity of the interface position to the heater and cooler temperatures were greater for small Biot number \( \text{Bi} = hR/k \), where \( h \) = heat transfer coefficient, \( R \) = ampoule radius, and \( k \) = thermal conductivity, i.e. for small ampoule diameters, ineffective heat transfer from the ampoule surface and high thermal conductivity. For higher crystal growth rates, a tendency towards the formation of concave isotherms was observed[64].

In an extension of Chang and Wilcox's model, Fu and Wilcox[66] investigated a three zone furnace in which the hot and cold zones were separated by an insulation layer (adiabatic zone). All the assumptions used in Chang and Wilcox's model were made except for the use of unequal heat transfer coefficients in the hot and cold zones. Using a finite difference computer modelling approach, it was shown that the use of an adiabatic zone significantly reduced the curvature of the isotherms between the hot and cold zones and allowed much better control over the shape of the melt interface. However, this benefit was achieved at the expense of a loss in the axial temperature gradient needed to suppress morphological instabilities.

Naumann[69] developed a hybrid analytical/numerical model for the two dimensional heat flow in a hot zone/adiabatic zone/cold zone Bridgman configuration. An analytical solution was obtained for the temperature field in each zone and Fourier-Bessel coefficients were obtained numerically by solving a system of linear equations determined
by the boundary conditions at the zone interfaces. The two dimensional model accommodated different heat transfer coefficients in the hot and cold zones as well as different properties in the solid and liquid phases, and determined the position and the shape of the solidification isotherm.

Naumann and Lehockzy[72] concluded from an analytical study that large radial temperature gradients could exist near the growth interface within an insulating zone if the melt and crystal thermal conductivities were significantly different (e.g. for Hg_{1-x}Cd_xTe, \( k_l/k_s = 7 \)) and the ampoule carried a bulk of the heat flux (i.e. ampoule thermal conductivity much greater than the crystal conductivity). Using a finite element method, Chin and Carlson[73] showed that the interfacial curvature was strongly influenced by the solid-liquid thermal conductivity ratio, dimensionless interface temperature and furnace Biot numbers and to a much lesser degree by the insulation zone thickness and ampoule position. It was noted that for meaningful results to be obtained from predictions, the material properties in the two phases should be accurately known.

Effects of melt convection on the interface shape and dopant segregation in the crystal were first investigated by Chang and Brown[74] using finite element methods which simultaneously solved for the velocity field in the melt, the shape of the solidification isotherm, and temperature distribution in both phases for gallium doped germanium. They concluded that convective fluid flows had a significant effect on radial dopant segregation. The length of the adiabatic zone and the ratio of thermal conductivities between the melt and crystal were identified as important parameters for determining the interface shape and the degree of radial segregation.

Numerical experiments were used by Carlson et al. [75] to study thermally driven flows. A parabolic representation of the solid-liquid interface was used to study convective flow patterns for various insulation thicknesses, Grashof, Prandtl and Biot numbers. For convex interfaces, a single cell was found and for concave interface shapes, multiple counter-rotating cells were predicted. Kim and Brown[91,92] employed two-dimensional, axisymmetric finite element models to study heat transfer, convection, segregation, and the
dynamics of HgCdTe growth. The large thermal conductivity ratio between the liquid and solid (≈7) resulted in highly concave interfaces.

Only a few modelling studies have focused on CdTe and Cd$_{1-y}$Zn$_y$Te alloys. Sen et al. [14] considered a quasi-steady state two-dimensional axisymmetric model for an ampoule containing the charge (a 2 inch diameter sample). They included conductive and radiative modes of heat transfer but neglected latent heat effects and convective fluid flow. Using a finite element analysis code (ANSYS), a heat transfer analysis was performed to calculate isotherms and find the position and curvature of the solid-liquid interface.

Pfeiffer and Muhlberg [53] also used a finite element simulation to calculate the shape of the interface. In addition to conduction and radiation effects, the release of latent heat was also included in the analysis. However, convection was still neglected, and growth rate was assumed to be equal to the ampoule lowering rate. The interface shape was reported to be initially convex in the first-to-freeze region due to heat loss at the ingot tip. In the intermediate region of the ingot, the interface was reported to be predominantly influenced by the ratio of the thermal conductivities of the solid and liquid phases of CdTe (concave). In the last-to-freeze region, the interface shape became progressively convex due to the lower heat input and the heat loss through the top region. Two sample diameters (27mm and 7.5mm) with two different aspect ratios (diameter/length) were considered. The lengthening of the ingot was shown to yield more single crystalline material (reduced end effects).

Parfeniuk et al. [10] calculated temperature and stress fields from a transient finite element model formulation. Latent heat release at the interface was incorporated in the model. Convection was neglected. For low axial temperature gradients (≈10 K/cm), a relatively flat solid-liquid interface was obtained. Increasing the furnace gradient above 10 K/cm changed the interface to a convex shape. Lowering the gradient (below 10 K/cm) promoted a concave interface shape. For the temperature gradients which produced convex interface shapes, the growth velocity was found to have a negligible effect on the interface curvature.
Recently, Kuppurao et al. [30,42-44] analyzed the growth of 75mm diameter zinc doped cadmium telluride using massively parallelized finite element models run on supercomputing platforms. These accounted for heat transfer, melt convection and zinc segregation. A two-dimensional, quasi-steady-state model was used to study the effects of heat transfer and melt convection[42]. Large radial gradients were shown to dominate the temperature in the solid, while convection flattened the radial temperature distribution in the melt. Concave interface shapes were predicted and the shape of the solid-liquid interface was found to be sensitive to the growth rate due to the importance of latent heat release. A transient growth analysis was used to investigate the segregation of zinc[30]. It was observed that significant axial and radial segregation was produced by convective mixing in the melt. Lowering the growth rate was predicted to slightly increase axial segregation but reduce radial segregation.

A second domain where numerical simulation has provided a better physical understanding of crystal growth processes is the calculation of stresses in melt grown crystals[96-105]. Stress is a common problem in grown crystals. Dislocations can form as a direct result of thermal stresses. The prediction of the dislocation density in the growing crystal is of key interest to the crystal grower since it provides a measure of the final crystal quality.

Stress is introduced during the extraction of heat from the crystal surface. The control of stress in the cooling crystal is one of the most important factors with respect to the final crystal quality. The stresses result from the temperature gradients which exist in the crystal. In addition, stresses can also be introduced by the agglomeration of point defects, inhomogeneous stoichiometric composition or inclusions. Very little is known about the elastic behavior of single crystals at elevated temperatures or the material properties when approaching the melting point.

Billig observed that the dislocation density of germanium crystals was correlated to the imposed temperature gradient [96]. Jordan et al. [97] made a first attempt toward a
quantitative description of the relationship between thermal conditions, thermally induced stress and resulting dislocation density for Czochralski grown crystals.

Huang et al. [98] used finite element analysis to calculate the stress distribution in crystals grown by the Bridgman method. It was found that the temperature distribution, and particularly the changes in temperature gradient, were the main factors governing the maximum thermally induced stress. The maximum stress was found to be located around the outer surface of the crystal. The solid-liquid interface shape was reported to be a secondary factor.

Rosch and Carlson [99] approximated the thermoelastic stress field of GaAs crystals grown by the vertical Bridgman method using a linear elastic, axisymmetric stress model. The model included effects due to the thermal field and the gravitational field, as well as the interaction with the ampoule, but did not account for the elastic anisotropy of the crystal. The difference between the Von Mises stress and the critical resolved shear stress (CRSS) was used as a measure of the number of dislocations present. The crystal-ampoule interaction was found to be the most important parameter in dislocation generation. Concave interfaces were predicted to have lower stress than convex interfaces.

In the thermoelastic models, the use of elastic modulus to convert thermal strains to stresses and the association of dislocation generation and its movement with a critical resolved shear stress are approximations. Dislocation movement is indicative of plastic flow. Volkl and Muller [100] developed a model which treat plastic deformation as a dynamic process, taking into account the generation and multiplication of dislocations. The model considers the relaxation of the thermally induced stress in the crystal, the strain hardening by generated dislocations, the variation of the stress level and the temperature variations during the whole growth process.

The validity and comparison of models of the crystal growth processes depend on the effect of simplifying assumptions on the different mechanisms and the accuracy of thermophysical properties used in each model. Often, thermophysical data at elevated
temperatures are not available. Further, supercooling of the melt and growth from non-stoichiometric melts are not modelled. Sensing approaches can serve purposes by validating available theoretical models, providing insights into the growth process, enabling on-line feedback control, and sometimes providing measurements of physical properties. Chapter 3 now presents a review of different sensing techniques that have been explored for crystal growth technology.
Chapter 3: Survey of Interface Measurement Methods

While destructive techniques such as quenching (by interrupting growth cycles) and vibrational methods have been usually utilized to identify interface locations/shapes, they involve post-processing of the grown crystal in order to make the interface demarcation lines visible. However, there are many physical differences between the solid and liquid phases of semiconductors near their melting temperatures. Any could be considered a basis for non-destructive sensing of interface position and shape. They include (together with their measurement methodologies): refractive index (optical scattering), density (x- or γ-ray radiography/tomography), elastic moduli (ultrasonic imaging), emissivity (infrared imaging), and electrical conductivity (eddy current measurements). The selection of an appropriate sensing method would depend on the sensitivity of the sensor response to an interface position and/or shape measurement and also the capability of its usage in a hostile, high temperature environment with minimum perturbations to the growth conditions.

3.1 Quenching

Capper et al. [119] used quenching as a technique to reveal the interface shape (at the time of quenching) in Hg$_{1-x}$Cd$_x$Te crystals grown by the Bridgman process. In some cases, a single quench into air was used to produce an interface in a specific part of the crystal and in others a multi-quench was performed. The multi-quench approach enabled them to study effects of sections of rapid growth on the crystallinity of the material by comparing the position and shape of the interface at various points during growth. In all cases, the quenched interfaces were found to be approximately parabolic in shape and concave with respect to the solid. A qualitative correlation between the interfaces and the radial compositional variations (determined by infrared transmission measurements) was observed.

In another similar study, Huang et al. [120] investigated the melt-solid interfaces obtained during vertical Bridgman growth of (Pb,Sn)Te crystals by a rapid quenching in
water and a subsequent electrochemical etching of longitudinally cut sections. The shapes of the interface shape were correlated to the composition variations determined by electron microprobe analysis. The composition variations along the growth axis indicated strong convective mixing in the melt. The experiments also demonstrated that the interface shape could be changed from concave to convex by moving its location from the edge of the cold zone into the hot zone.

In order to make visible the solid-liquid interface, Pfeiffer and Muhlberg[53] used a slightly Te-rich (3-4 at%) CdTe melt to cause a sudden breakdown of stable growth conditions by turning off the electric power to the furnace zones and rapidly lowering the ampoule in the low-temperature region of the vertical Bridgman furnace. The interface was observed to be initially convex in the first-to-freeze region due to heat loss at the ingot tip. In the intermediate region, the interface was found to be concave because the thermal conductivity ratio \( k_s/k_l \) was less than 1. Similar to the first-to-freeze region, the last-to-freeze region was noted to be influenced by end effects (heat loss through the top region). In a slight variation of Pfeiffer's method, Dutta et al. [121] performed the growth, power shut down and quench sequence twice for GaSb crystal growth runs. After the completion of the growth run, crystals were cut along the growth axis and chemically etched. Knowing the distance between two subsequent demarked boundaries, and the time in between the two quench segments, the growth rate was evaluated.

3.2 Vibration methods

In general, vibrational interference should be avoided in order to retain high perfection in the growing crystal. Although various types of insulating measures are taken, it is difficult to prevent the growth of crystals being affected by external vibration. Vibrations of known frequency introduced into a melt during crystal growth appear as impurity "striations" in the growing crystal. Witt and Gatos [122,123] used this observation to develop a technique for determining the instantaneous microscopic growth
rates and interface morphology of Czochralski grown InSb single crystals (Figure 3.1). Small amounts of tellurium were introduced in the melt as the detecting impurity.

High frequency low amplitude vibrations were introduced by coupling (contacting) a vibrating rod to the crucible containing the melt. A signal generator and an audio amplifier were used as the source of the known excitation frequencies. From the known frequency of the vibrations, and spacing of the resulting striations, the microscopic rates of growth were found. By using suitable etching techniques and interference contrast or dark field microscopy, striations 0.2μm apart were distinguished. The crystal-melt interface was determined from the direction of the vibrational striations. In a similar study, Quang et al. [124] investigated the influence of mechanical vibrations on microscopic growth rates in GaSb crystals pulled from the melt.

Figure 3.1 Schematic representation of Czochralski InSb crystal growth arrangement with induced vibrations [123].
3.3 Radiography

Semiconductors at their growth temperature, are opaque to visible light, and Bridgman furnaces are also constructed from materials which are usually opaque. To penetrate the furnace and sample, the use of X-rays and (higher energy) γ-rays was proposed by researchers at NASA Langley in 1986 as a real-time imaging technique for in-situ determination of solid-liquid interface shapes during Bridgman growth[125]. These techniques were proposed because of the large differences in densities of the crystal and melt. This results in different image intensities in either a X- or γ-ray image. Two semiconductor materials were tested during the development of this method. Germanium was selected because of its relatively low molecular weight and low atomic number, thus penetrable by X-rays. Lead tin telluride (Pb,Sn)Te was chosen as the second test material because it represented a ternary compound semiconductor which contained elements, especially lead, with high atomic number, and required penetration by gamma rays. A density difference of about two percent was considered to be a minimum required to be observed on high contrast X-ray film. Calculations prior to the experiments had revealed that for Ge and (Pb,Sn)Te, the density difference between the melt and the solid were around 4% and 10% respectively.

Initially, the time required to determine the interface position was considered too long (because of the long exposure time) for continuous monitoring. However, over the years, the quality of images were improved by incorporating image enhancement techniques. These now enable in-situ, real-time images of the interface position and shape to be obtained[126]. With present day technology, each image takes around 10s to acquire, which at a growth rate of 1 cm/h results in only 28μm of growth between two consecutive images. In a technological development very similar to this, Kakimoto et al. [127,128] observed fluid flows and interfaces in Silicon grown in a Czochralski furnace (Figure 3.2).

Barber et al. [129] used radiographic technology to directly observe the melt-solid interface position and shape during crystal growth of germanium and lead tin telluride. They determined the growth rates for the two material systems and found that for
germanium, the growth rate exceeded the furnace translation rate. Over the course of the experiment, the melt-solid interface in germanium changed its position with respect to the furnace, gradually moving toward the upper, hot zone. As the interface migrated from near the cold zone to near the hot zone in the Bridgman furnace, the interface shape changed from concave to convex. On the other hand, the growth rate for the compound semiconductor (Pb,Sn)Te was less than the translation rate, with the melt-solid interface position gradually moving down toward the cold zone of the furnace.

Figure 3.2 Schematic diagram of Czochralski crystal growth chamber with a X-ray radiography system[128].

In a recent study, Campbell and Koster [130,131] demonstrated a radiographic technique for real-time visualization of the melt/crystal interface of indium antimonide. Appropriate furnace materials were selected that were relatively transparent to X-rays for proper imaging. For indium antimonide the melt is 10.3% more dense than the solid at the
melting point, and hence the x-ray absorption of the melt is higher than that of the solid. The penetrating radiation energy was supplied by a 160kV x-ray source. The x-ray source and detector were maintained in a fixed position with respect to the ampoule while the furnace translated. Image processing techniques were used to enhance the contrast between the more absorbing melt and less absorbing crystal. The shape of the interface was observed to be slightly concave.

3.4 Ultrasonic methods

Ultrasonic pulse-echo techniques have been used to locate the solid/liquid interface during solidification and melting of metals, alloys and semiconductors in Bridgman furnaces[132,133]. These techniques rely on an acoustical impedance difference between solid and liquid phases. In the pulse-echo technique, a piezoelectric transducer is used to send and receive an ultrasonic pulse. Solid/liquid interface detection is made possible by a measurable difference in both sound velocity and density across the interface which is indicated by the partial reflection of a sound wave at the interface. Solid/liquid interfaces in low-melting pure substances have been satisfactorily located but results with the much higher melting iron alloys were less encouraging due to a low signal/noise ratio attributed to no identifiable reflection from the “mushy zone”. Most metals exhibit a reduction in both velocity and density during a solid to liquid transition, and exhibit a small (~20%) velocity change. Semiconductors, on the other hand usually become more dense (in the liquid state) and therefore show a larger reduction in velocity. The success of the method depends on the degree of reflection of the signal. Higher reflection coefficients, \( R \), given by \( R = \frac{Z_s - Z_l}{Z_s + Z_l} \) (where \( Z_s \) and \( Z_l \) are the acoustic impedances of the solid and liquid phases respectively) are typical of semiconductor materials (compared to metals) which could be taken as an indicator that this method would be an appropriate method for interface detection of semiconductors. Jen et al. [134] demonstrated the use of the pulse-echo method for monitoring the float zone crystal growth of Ge. It was shown that arrival times of the reflected signals from the solid-liquid Ge interface could be used to determine the position of the interface.
When the echo signal is weak and difficult to detect, through transmission time-of-flight measurements provide an alternate way of interrogating an interface exploiting refracted rays. The time of flight for a ray traversing a solidifying body depends upon the fractions of the ray's path within the liquid and solid phases and the velocity within each phase. Carter et al. [135] propagated ultrasound parallel to the growth direction of polycrystalline Germanium. They were able to locate the interface within 0.3mm but could not reveal information on the interface shape.

The ultrasound methods described above used piezoelectric transducers to generate and detect sound waves. The use of piezoelectric transducers however is restricted by the application temperature of the sensor and the ability to maintain a good mechanical contact with the sample at elevated temperatures through an appropriate acoustic coupling medium. Laser based ultrasound sensing has emerged as an attractive choice for applications which require non-contact measurements in high temperature materials, and operation in hostile environments [136-139]. It has been successfully used in the determination of ultrasonic velocities and elastic constants in metals, ceramics, composites, and semiconducting compounds both at ambient and elevated temperatures. The ultrasonic velocity in pure, single phase, isotropic materials has almost a unique dependency on temperature. Measurements of time of flight have been used to predict temperature fields and interface position in metals. Ultrasonic time-of-flight data has also been used in reconstructing the two dimensional boundary of a liquid core in a solidifying strand of aluminum[137].

In single crystal growth applications, non-contact laser generation and detection of ultrasound has been used in the optimization of Czochralski growth of metals and semiconductors. A pulsed laser beam was used to generate an elastic wave on one side of a single crystal during pulling from the melt, while a continuous laser interferometric detector, incident on the opposite side of the crystal was used to record the transit time of the wave. Investigations have also been undertaken to explore the recovery of the interface position and shape of vertical Bridgman grown Cd$_{1-x}$Zn$_x$Te [138,139]. Here, a laser
ultrasonic method was used with a model system with ultrasonic velocities similar to the liquid and solid velocities of Cd$_{1-x}$Zn$_x$Te (Figure 3.3). A ray tracing code was developed and used to predict ultrasonic ray paths and time of flight as a function of axial position for a variety of interface shapes.

![Figure 3.3 Schematic of laser-ultrasound measurement system][139].

### 3.5 Eddy current sensing

Each of the methods described above has limitations for characterizing the solid-liquid interface of a growing semiconductor crystal. The quenching and vibration methods are destructive to the crystal. The x-ray (or γ-ray) radiography and laser based ultrasound methods are expensive, have certain safety hazards and require modifications/re-design of the furnace.

Eddy current sensors operate on the principle of electromagnetic induction and are non-contacting and potentially non-invasive (provided the eddy current density remains low). They are inexpensive, introduce no safety hazards and can be used in ways that
minimally perturb crystal growth environments. They employ time-varying electromagnetic fields as a probing medium to explore, (1) the electromagnetic properties of a test material (i.e. the electrical conductivity, \( \sigma \), and the magnetic permeability, \( \mu \)), (2) the variations in geometry and dimensions of a test specimen (e.g. thickness, diameter), and (3) the existence of discontinuities (e.g. cracks). The time varying electromagnetic fields which are usually created by alternating current flow in a (excitation) coil, induce the flow of electric currents (i.e. eddy currents) within a nearby conducting medium. Specimen defects, changes in electrical conductivity and/or magnetic permeability, dimensional variations affect the flow of eddy currents which can all be measured as a change in impedance of the excitation coil or a nearby search (pickup) coil. Sometimes, the sensor output is analyzed in terms of the magnitude and/or phase angle response of the magnetic vector potential in the sensed region.

Most semiconductors exhibit an abrupt decrease in electrical conductivity at the liquid to solid transition during solidification, thereby providing an incentive for using eddy currents to detect the solid-liquid interface. Wallace et al. [140], Tien et al. [141], Stefani et al. [142,143] and Choe et al. [144-146] conducted a variety of eddy current sensing experiments with 70-80mm diameter Czochralski grown Silicon crystals to map out the thermal field. A single turn coil was used as the excitation and detection coil (Figure 3.4). In this arrangement, the coil sensed small sample induced perturbations to its own field. Axial and radial temperature profiles were calculated from the eddy current sensor conductivity data using a predetermined temperature-electrical conductivity relationship for Silicon.

A two coil eddy current sensor arrangement was used in a model study of Czochralski grown 76mm diameter GaAs crystals by Wadley et al. [115] to investigate effects of interface shape and melt height. The responses of several pickup coil configurations to a multiple turn excitation coil were analyzed to identify appropriate designs which enhanced the sensitivity to melt height or interface shape change.
Figure 3.4 Schematic diagram of a single coil eddy current sensor used for monitoring Czochralski grown Silicon crystals.

Larkin et al. [147] performed an interface simulation test of the vertical Bridgman process using a graphite-liquid gallium model system. Three graphite cylindrical samples with one end machined to a prescribed shape (convex, flat, concave) were placed inside a plastic tube and liquid gallium filled on top to emulate a melt-crystal interface. (Figure 3.5). An encircling eddy current probe was scanned vertically to obtain the phase angle response of the magnetic vector potential. A scheme which compared the peak position of the first derivative of the phase angle response relative to the mean of the phase angle was proposed to determine the interface shape (Figure 3.6). No mention was made whether the selection of test frequency had an influence on the analysis protocol, and if so, no systematic method of choosing the appropriate frequencies was presented.
Figure 3.5 Schematic diagram of the interface simulation test[147].

Figure 3.6 Phase angle response during eddy current sensor translation[147].
Successful applications of eddy current sensing to interface characterization rely on knowledge of the electrical conductivity-temperature relationship near its melting point. For CdTe, the data available in the literature is limited to previous investigations by Glazov et al. [108]. The accuracy of this data has been questioned due to a lower melting point temperature reported in their study. Rosen et al. [148,149] conducted experiments with indium-doped cadmium telluride using a single coil eddy current sensor design for electrical conductivity measurements. In addition, the single coil sensor was excited at 4 frequencies to monitor the amplitude and phase variations during growth from a melt of 50mm diameter crystals in a 2-zone vertical Bridgman furnace. Recently Choi [150,151] used a multifrequency eddy current sensor to measure the electrical conductivity of Cd$_{1-x}$Zn$_x$Te alloys (for x=0, 0.45, and 0.8) as a function of temperature during heating and cooling through the melting transition. A 4-6 fold increase of conductivity was observed during the solid to liquid phase change. The presence of Zn was noted to decrease the liquid conductivity.

Ideally, sensors for crystal growth should operate in a non-contact configuration with minimum perturbation of the growth process. In addition to characterizing the interface, sensors that can provide information on the melt composition and temperature fields in the melt and crystal are beneficial to the crystal grower. The eddy current sensing approach looks the most promising in meeting these sensing requirements. In the application of eddy current sensors to Bridgman crystal growth, the signals from the pickup (or search) coil vary with time depending on the changing position and shape of the interface and the electrical conductivities across the interface. The electromagnetic interactions between the sensor and the changing conditions of the sample in the vicinity of the sensor are complex. Experimental studies have been limited in scope with few systematic evaluations of the results. There has also been a lack of data from real growth runs.

In this research, electromagnetic finite element methods have been applied to simulate the responses of eddy current sensors interrogating solidifying crystals. Bench
top tests were then done to validate these models. Protocols to distinguish interface position and shape have been developed based on the electromagnetic skin depth differences in the solid and liquid. An in-situ high temperature sensor was integrated into a commercial scale crystal grower. In these investigations, the eddy current sensor has demonstrated the ability to provide useful information on growth conditions and a better understanding of the crystal growth process. The potential now exists to use the sensor to also control the crystal growth process.
Chapter 4: Eddy Current Sensor Theory

4.1 Introduction

The eddy current sensing technique is an electromagnetic method of nondestructive evaluation which employs time-varying electromagnetic fields as a probe to explore, (1) the electromagnetic properties of a test material (i.e. the electrical conductivity, $\sigma$, and the magnetic permeability, $\mu$), (2) the variations in geometry and physical dimensions of a test specimen (e.g. thickness, diameter), and (3) the existence of discontinuities (e.g. cracks, porosities, inclusions)[152]. The varying electromagnetic fields are usually created by the flow of a periodic (sinusoidal or pulse) current through an excitation or "driver" coil. The magnetic field associated with this excitation current induces a flow of electric currents (i.e. eddy currents) in a nearby conducting sample. These follow closed paths in planes perpendicular to the direction of the magnetic field and in a direction opposite to that of the primary coil current (in accordance with Lenz's law). The current distribution in the sample is determined by the electrical/magnetic properties of the test sample and by the geometry of the coil system and sample. The induced eddy currents in the sample generate a secondary field which interacts with the primary magnetic field. The effect of this perturbation is typically measured as a change in electrical impedance (magnitude and phase) of the excitation coil (in a single coil system, Figure 4.1) or as a "transfer" impedance if a separate "search" (or pickup) coil is used in a two coil arrangement, Figure 4.2.

![Figure 4.1 A single coil eddy current sensor encircling a cylindrical sample.](image-url)
Figure 4.2 A two coil eddy current sensor arrangement for transfer impedance measurements consisting of a primary coil providing the excitation current and a secondary coil to pick up the induced voltage.

Much like acoustic waves in ultrasonic testing or x-rays in radiography, eddy current testing involves the transmission of energy in the test specimen. In the eddy current case, the probing radiation is electromagnetic with a frequency in the radio part of the electromagnetic spectrum.

The selection of the test frequency determines the penetration depth of fields into a sample. It is often characterized by an exponential decay of a plane wave field's penetration into a planar test material. The depth of penetration at which the field intensity falls to a value 1/e (=0.368) from that at the sample surface (often referred to as the "skin depth", $\delta$) is given by,

$$\delta = \sqrt{\frac{2}{\omega \mu \sigma}}$$  

where, $\omega$ is the test frequency, $\mu$ the magnetic permeability and $\sigma$ the electrical conductivity. This is referred to as the "skin effect" phenomenon, where eddy currents in conducting media tend to be concentrated near the surface adjacent to the excitation coil. Increasing the operating frequency, sample electrical conductivity and magnetic permeability increases the "skin effect" resulting in shallower depths of penetration of the field into the sample.
The eddy current density at any depth $x$ from the surface is given by,

$$J_x = J_0 \exp \left( \frac{x}{\delta} \right)$$  \hspace{1cm} (4.2)

where, $J_0$ is the current density at the surface. The skin effect is schematically illustrated in Figure 4.3.

![Diagram of eddy currents](image)

Figure 4.3 Relative effect of test frequency, electrical conductivity, and magnetic permeability on depth of penetration.

Part of the energy transmitted is absorbed in the test specimen and dissipated as heat (resistance heating). Depending upon the electrical properties, internal interfaces, and the surface condition of the test specimen, the remaining energy is reflected. The net result is that the apparent impedance of the excitation (primary) or pickup (secondary) coil changes in the proximity of any conducting medium. The changes are related to the sample's geometry and electrical properties, the test coil design and the test frequency.

Normally, the impedance of the test coils is complex and changes with frequency. A trace or locus of the (complex) coil impedance plotted with the coil resistance on the $x$-axis and the inductive reactance on the $y$-axis is referred to as the "impedance plane
diagram" and is commonly used to display eddy current test data. Often, the impedance measured with a sample present is normalized with respect to an empty coil measurement. This tends to emphasize the relationship of the data to the sample properties and geometry. The solid line in Figure 4.4 shows a characteristic normalized impedance diagram for a coil of inner diameter, $D_0$, encircling a cylindrical sample of the same diameter, $D_0$.

For cylindrical configurations, the ratio of the sample cross-sectional area to the pickup coil area is defined as the fill factor[152,153]. As the fill-factor decreases (i.e. the sample diameter decreases relative to the coil diameter), the impedance curve moves to positions indicated by the short dashed lines (Figure 4.4). This shift is utilized for in-situ measurements of dimensional changes during consolidation processing[154,155] since the high frequency intercept on the normalized impedance curve is equal to (1- fill factor), providing a convenient way of measuring the changing sample diameter.

![Normalized Impedance Diagram](image)

**Figure 4.4** A characteristic normalized impedance curve for a coil encircling a solid cylinder.
Increasing the operating frequency shifts the data on the impedance curve in the clockwise direction, while an increase in the electrical resistivity of the sample causes a shift in the opposite direction. In applying the eddy current sensing method to Bridgman crystal growth, it is this latter property (i.e. the impedance change due to sample resistivity variations) that one seeks to exploit since, most semiconductors exhibit a considerable decrease in the electrical conductivity during the liquid-solid phase change [108,150,151].

For crystal growth applications, a single coil system provides the simplest sensing arrangement [140-149]. However, a two coil system is preferred in high temperature applications since a transfer impedance measurement allows the induced voltage in the secondary coil to be measured with a high input impedance instrument which minimizes temperature dependent resistance changes from affecting the impedance measurements. Furthermore, a two coil arrangement provides the flexibility for evaluating alternate pickup coil arrangements. This may lead to designs that show increased sensitivity to a moving liquid-solid interface (e.g. "absolute" vs. "differential" sensor pickup coils, Figure 4.5. In the two coil eddy current sensor system, the transfer impedance is calculated from the induced voltage in the secondary for a known current flowing through the primary. In section 4.4, expressions for the induced voltage are derived starting from the point form (differential) representation of Maxwell's equations in section 4.2.

![Figure 4.5 Pickup coil arrangements. a) an "absolute" sensor. b) a "differential" sensor](image-url)
4.2 Electromagnetic field equations

All electromagnetic phenomena, including those related to eddy current sensing are described by Maxwell’s equations. The differential equations governing time varying fields in regions that include conducting materials can be derived from Faraday’s, Ampere’s and Gauss’s laws and the divergence theorem.

Faraday’s law:

$$\nabla \times E = \frac{\partial B}{\partial t}$$  \hspace{1cm} (4.3)

Ampere’s law:

$$\nabla \times H = J + \frac{\partial D}{\partial t}$$  \hspace{1cm} (4.4)

Gauss’s law:

$$\nabla \cdot D = \rho$$  \hspace{1cm} (4.5)

$$\nabla \cdot B = 0$$  \hspace{1cm} (4.6)

where, $E$ and $H$ are the electric and magnetic field intensities, $B$ and $D$ are the magnetic and electric flux densities, $J$ is the current density and $\rho$ the charge density. The conduction current term $J$, for metals and semiconductors is usually much larger than the displacement current term $\frac{\partial D}{\partial t}$ in the frequency range that eddy current sensors are operated. Hence the $\frac{\partial D}{\partial t}$ term in equation (4.4) can be ignored.

In addition to Maxwell’s equations, the simple constitutive relations describing the material media can be used.

$$B = \mu H$$  \hspace{1cm} (4.7)

$$D = \varepsilon E$$  \hspace{1cm} (4.8)

where $\mu$ is the magnetic permeability and $\varepsilon$ the dielectric constant.
The current density, $J$, is related to the electric field intensity $E$ by a form of Ohm's law,

$$J = \sigma E$$  \hspace{1cm} (4.9)

where $\sigma$ is the electrical conductivity.

The magnetic permeability, $\mu$, the dielectric constant $\epsilon$, and the electrical conductivity, $\sigma$ for anisotropic media require 3-D tensor representation. For a linear, isotropic medium, scalar constants can be used in equations (4.7), (4.8), and (4.9) assuming independence from the field.

The magnetic flux density $B$ can be expressed as the curl of a vector $A$ (commonly referred to as the Magnetic Vector Potential),

$$B = \nabla \times A$$  \hspace{1cm} (4.10)

Substituting for terms in equations (4.7) and (4.10) in equation (4.4) we get,

$$\frac{1}{\mu} \cdot \nabla \times (\nabla \times A) = J$$  \hspace{1cm} (4.11)

Using the vector identity,

$$\nabla \times (\nabla \times A) = \nabla (\nabla \cdot A) - \nabla^2 A$$  \hspace{1cm} (4.12)

and the Coulomb gage condition,

$$\nabla \cdot A = 0$$  \hspace{1cm} (4.13)

equation (4.11) reduces to,

$$\nabla^2 A = -\mu J$$  \hspace{1cm} (4.14)

where $J$ consists of the source current ($J_s$) and eddy current densities ($J_e$) given by the relationship,

$$J = J_s + J_e$$  \hspace{1cm} (4.15)
With sinusoidal excitation (of frequency \( \omega \)), equations (4.14) and (4.15) reduce to the eddy current diffusion equation,

\[
\nabla^2 A + j\omega \mu \sigma A = -\mu J_z 
\]

(4.16)

The boundary conditions are that the vector potential, \( A \) and its normal derivative \( \frac{\partial A}{\partial n} \) are continuous across each material interface. Equation (4.16) represents a partial differential equation for the eddy current problem which can be solved with an analytic scheme or (more commonly) with a numerical method.

### 4.3 Analytical methods

Analytical methods to solve for phenomena associated with eddy current sensing (e.g. the calculation of the induced voltage or coil impedance) involve the simplification and manipulation of Maxwell's equations into forms where standard mathematical methods such as the separation of variables and Fourier Transforms can be applied. In most cases, a governing second order partial differential equation is first derived in either the flux density or vector potential form. In general, problems are tractable only if simplifying assumptions are made concerning both material properties and test geometry.

Forster and Stambke [156] investigated the effects of a metal rod encircled by an AC excitation coil and a secondary search coil using Bessel functions. Hochschild [157] used Bessel functions to analyze the case of a cylindrical sample and a concentric coil in order to calculate the flux density within the conductor in terms of the flux density at the conductor's surface. An expression for the flux linkage with the encircling coil was derived from which the induced voltage and the real and imaginary parts of the coil impedance were found. Libby[153] used a magnetic vector potential representation of Hochschild's geometry. Dodd and Deeds obtained expressions for the impedance of a coil encircling a two conductor rod [158]. Magnetic vector potential solutions were obtained in terms of integrals of Bessel functions.
Linear, isotropic, and homogeneous cylindrical geometries were assumed in each of the above analyses. The major drawback of the analytical methods is the lack of generality. Solutions are restricted to a limited class of problems involving basically homogeneous and linear media with simple geometric configurations.

In vertical Bridgman crystal growth, although the sample preserves an external cylindrical geometry (defined by the ampoule or crucible diameter), the "effective" cylinder is non-homogenous since it consists of two phases (solid and liquid) of the material which have different electrical conductivities. Since the interface shape is not known a priori, one should be able to analyze the changes in impedance responses for a variety of interface shapes (convex, flat, concave). In addition, since the solid-liquid fraction "sampled" by the sensor varies with the movement of the interface, the analysis tool should be flexible enough to cover the range of possibilities (position and shape) likely to be encountered during a Bridgman growth run. From this point of view, numerical methods have a distinct advantage over the more rigid analytical solution schemes.

4.4 Numerical methods

4.4.1. Introduction

Numerical methods are based on either integral or differential equations. Though physically equivalent, each method has relative strengths and weaknesses due to numerical limitations. They are both applied by discretization of the problem domain and the derivation of a system of equations whose solution give the numerically computed approximate field. The boundary element method is based on the boundary integral equivalent of the governing differential equation using Green's functions. They excel with open boundary problems. Far fewer unknowns are involved in integral methods, but the matrices derived are full, sometimes leading to considerable numerical difficulties. The main advantages of the differential methods (e.g. finite difference, finite element) are in handling complicated geometries and materials properties. The matrices derived for
differential methods are sparse and banded allowing very efficient matrix solution methods to be used. The differential methods have matured rapidly and several convenient codes are available for solving problems.

In the finite difference method, a discretization procedure is followed whereby the differential equation describing a given problem is converted into a finite-difference equation. A grid is superposed in the problem domain where the value of the field variable at a node is related to the values of its neighboring nodes. This method has been used in the prediction of electromagnetic fields in electrical machinery. It is more suitable for regular domains, where an orthogonal mesh (in rectilinear or polar coordinates) can be conveniently used. They do not conform easily to complicated irregular surfaces (e.g. a more complex solid-liquid interface) which can be treated by a finite element mesh.

The electromagnetic finite element method is based on variational calculus[159,160]. Rather than solving the eddy current diffusion equation directly by representing the partial derivatives with a difference equation, the finite element approach seeks to minimize an energy functional over the discretized region of interest yielding values of the field variable (say, the magnetic vector potential, $A$) at every node of a chosen mesh. Once the vector potential is obtained, the induced voltage and/or the impedance can then be calculated.

For analyzing the vertical Bridgman geometry, the finite element method is preferred since conductivity variations and interface position and shape changes (complicated geometries) in the problem domain can be handled more easily.

4.4.2. The finite element approach

In the finite element method of electromagnetic analysis, one uses the variational formulation of the differential equation of the physical system in terms of an equivalent energy functional. For linear media and sinusoidal excitation, an energy functional ($F$) can be written;
\[ F = \left\{ \frac{B^2}{2\mu} - \frac{J \cdot A}{2} + \frac{j\omega \sigma}{2A^2} \right\} dV \]  \hspace{1cm} (4.17)

where \( J \) is the applied current density of angular frequency \( \omega \), \( \mu \) is the permeability, and \( \sigma \) the electrical conductivity. The three terms in equation (4.17) are the stored magnetic energy, input electrical energy and the dissipated energy respectively. A matrix equation is derived by minimization of the energy functional by setting its derivative equal to zero.

\[ \frac{dF}{dA} = 0 \]  \hspace{1cm} (4.18)

The resulting electromagnetic matrix equation over all grid points of a finite element model is,

\[ [M] \{ A \} = \{ J \} \]  \hspace{1cm} (4.19)

where, \([M]\) is a global matrix composed of the material properties (\( \mu \) and \( \sigma \)), the element geometry, and the test frequency \( \omega \), \( \{ A \} \) is the unknown vector potential, and \( \{ J \} \) is the known source current vector.

When the vector potential solution has been obtained, the induced voltage in the secondary coil can be calculated from the line integral of the electrical field intensity around the pickup coil circuit.

\[ V = \oint E \, dl \]  \hspace{1cm} (4.20)

From equations (4.3) and (4.10),

\[ E = \frac{\partial A}{\partial t} \]  \hspace{1cm} (4.21)

For sinusoidal excitation of frequency, \( \omega \), equations (4.20) and (4.21) can be combined to yield,

\[ V = -j\omega \oint A \, dl \]  \hspace{1cm} (4.22)
For axisymmetric geometries, equation (4.22) reduces further to,

$$V = -j\omega A_{ave}(N \cdot 2\pi r)$$ \hspace{1cm} (4.23)

where, $r$ is the radius of the pickup coil, $N$ is the number of turns, and $A_{ave}$ the average (complex) magnetic vector potential in the pickup coil region. The transfer impedance, $Z$, can then be calculated as,

$$Z = V/I$$ \hspace{1cm} (4.24)

where, $I$ is the known input current.

In chapter 5, an axisymmetric finite element model is developed to carry out simulations for the response of two eddy current sensor designs for several positions and shapes of a liquid-solid interface.
Chapter 5: Finite Element Model Simulations

5.1 Introduction

The finite element method has been used to evaluate the response of two eddy current sensor designs for a variety of liquid-solid interface locations/curvatures, and for several different materials systems. This allows quantitative relationships to be obtained between growth parameters such as the liquid-solid interface location/shape and measurable quantities of an eddy current sensor's response (e.g. the frequency dependent test coil impedance). This approach also has the advantage of allowing anomaly free protocols to be designed for deducing the growth parameters from measured experimental data, and provides guidelines for evaluating their potential for other material systems. Since both the absolute conductivity and the conductivity difference are likely to affect the performance of this sensing approach, the study explores the application of eddy current methods to a variety of semiconductors, (see Table 5.1). Silicon, though not commercially produced by a Bridgman method, is included in the materials analyzed to more fully span the conductivity range, and to better establish the sensor performance-test material conductivity relationship.

<table>
<thead>
<tr>
<th>Table 5.1: The electrical conductivities of selected solid ($\sigma_s$) and liquid ($\sigma_l$) semiconductors close to their melting point</th>
</tr>
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<tbody>
<tr>
<td>Semiconductor Material (S/m)</td>
</tr>
<tr>
<td>Electrical Conductivity (S/m)</td>
</tr>
<tr>
<td>$\sigma_s$</td>
</tr>
<tr>
<td>$\sigma_l$</td>
</tr>
<tr>
<td>$\sigma_l/\sigma_s$</td>
</tr>
</tbody>
</table>

Table 5.1 shows that for most semiconductors, the electrical conductivity of the liquid is many times that of the solid at the melting point. The principle underlying the application of an eddy current sensor approach to crystal growth is based on the
observation that the eddy current density induced at a point within a test sample by the electromagentic field of an a.c. excited coil is proportional to the sample's electrical conductivity at that point. Since the electrical conductivity of liquid semiconductors exceeds that of the solid, higher eddy current densities are expected to exist within liquid regions of a solidifying charge. Sensors based upon this principle have been previously proposed for measuring solidification conditions and temperature profiles during the Czochralski growth of GaAs and Silicon [115,140]. They are widely used in other types of high temperature materials processing, e.g. for determining internal temperatures within aluminum alloy extrusions [161,162] and for the measurement of dimensional changes during hot isostatic processing [154,155].

The response of an eddy current sensor is a complicated function of the eddy current distribution excited within the sample by the fluctuating electromagnetic field of an excitational coil. This will be affected by the geometry of the exciting coil (which governs the electromagnetic field's distribution within the test material), the coil's excitation current frequency, the fraction of material solidified in the interrogated volume, the shape of the boundary between the solid and liquid regions and the respective conductivities of the solid and liquid. In the eddy current technique, the distribution of eddy currents induced in the sample is sensed from their effect on the impedance of either the exciting coil or a separate "pickup" coil. It will be a sensitive function of the sensor's design and test frequency as well as all the material parameters listed above. Experimental methods might be used to perfect the sensor design, optimize the test frequency and develop data analysis protocols. However, it is costly and time consuming to equip a crystal grower with a variety of eddy current sensors, and to experimentally design a sensor approach. Furthermore, a definitive validation of the response is almost impossible because of the lack of independent observations of the solidification front.

Here, the simulated responses for two candidate eddy current sensor designs are obtained for each of the four material systems given in Table 5.1. The results are used to
investigate the effects of changing either the interface position or its shape on the sensor’s complex impedance.

5.2 Eddy current sensor designs

Figure 5.1 shows two encircling eddy current sensor designs selected for detailed study. Both have the same seven turn driver (primary) coil for excitation. One uses a single turn pickup coil located at the primary coil’s midpoint, while the other uses two opposingly wound pickup coils located near the ends of the primary coil. The single pickup coil sensor arrangement is called an “absolute” sensor (Figure 5.1a), while the two pickup coil sensor design will be referred to as a “differential” sensor (Figure 5.1b).

![Diagram of dual coil eddy current sensing arrangement]

Figure 5.1 Schematic diagram of a dual coil eddy current sensing arrangement. A seven turn driver coil is used to excite an electromagnetic field. Either a single coil or a pair of opposingly wound coils are used to “pick up” the perturbed flux.

For such a two coil sensor embodiment, the transfer impedance, $Z$, is given by

$$Z = \frac{V_s}{I_p}$$  \hspace{1cm} (5.1)

where $I_p$ is the excitation current in the primary (or driving) coil and $V_s$ is the voltage induced across the terminals of the secondary coil. The transfer impedance of such a
system is relatively insensitive to the resistance of the coils if, a) the induced voltage, $V_s$, is measured with a high impedance instrument and b) the current, $I_p$, is continuously monitored (e.g. by observing the voltage across a precision resistor placed in series with the primary coil). This enables operation of the sensor at high temperatures without the need either for correcting for coil temperature or (potentially invasive) coil cooling. With two coil systems like these, the primary coil’s axial length can also be made much longer than the secondary coil, so producing a relatively uniform field in the sensing region.

To envision the way such sensors might be used to monitor solidification during vertical Bridgman growth, consider a control volume element in the vicinity of either sensor (as indicated by the dashed areas in Figure 5.1). The length of this control volume is determined by the “effective” axial range of the electromagnetic flux created by the primary coil. Its penetration into the sample is controlled by the skin depth in the material under investigation. The induced eddy currents will be influenced by the volume fractions of the solid and liquid within this control volume. This changes if either the sample (containing a liquid-solid interface) remains stationary and the sensor is translated, or if the sensor/sample are both stationary and the growth furnace is translated (causing the solidification front to propagate along the sample). For this latter scenario, the region initially sampled would be liquid whereas at the end of growth (after the interface has moved through the control volume), the sensed region would be fully solid. It will be shown that the sensor’s response will always be bounded by its response to these two states, and all measured responses during growth must lie between these two extremes.

A single pickup (secondary) coil sensor design will be most responsive to the eddy current density closest to the secondary coil’s location. It is likely to be relatively unaffected by eddy currents excited far from the coil. The sensitivity of a sensor to the presence of an interface could potentially be improved by using a pair of opposing wound secondary coils located above and below an interface. The response of such a “differential” sensor will be dependent on the difference in sample created field perturbation at the two coil locations; common contributions to the two coils’ induction will be canceled out in this configuration. The sensitivity of a differential sensor measurement is likely to vary with the spacing between the two pickup coils providing an
additional degree of freedom for sensor design. The diameters of the primary and secondary coils were not considered as design variables. They are likely to be predetermined by the dimensions of the furnace geometry (e.g. liner tube) used for growth since this minimizes the perturbation of the thermal fields.

5.3 Finite Element Model

The problem modelled consisted of a cylindrical 76mm diameter sample containing one of five interface locations and five interface curvatures. Two of the interfaces had convex shapes (defined by a convexity parameter $\theta = z/D = +0.167, +0.333$ where $z$ is the interface curvature height on the axis and $D$ the test material diameter), one interface was flat ($\theta = 0.0$), and the remaining two were concave ($\theta = -0.167, -0.333$). The non-planar interfaces were hemispherical surfaces of differing radii of curvature. Since the sample encircled by the eddy current sensor was contained in a cylindrical crucible and the entire geometry was cylindrically symmetric, only one-half of an axisymmetric plane was analyzed, Figure 5.2(a). MSC/MAGGIE$^\text{TM}$[163] a personal computer based electromagnetic finite element software package was used to setup the model, generate the finite element mesh and run the simulations. It was assumed that the coil was circular symmetric (i.e. the helical effect of the coil was ignored). The primary coil was modelled as a series of circular loops of a known radius spaced a distance apart equal to the pitch of the primary coil winding. In addition, for axisymmetric problems no electromagnetic flux crosses the axis of symmetry, and hence a zero vector potential boundary condition can be specified on the axis.

In order to minimize the effects of mesh size on the solution, all five interface shapes were incorporated into one finite element model and the same automatically generated finite element mesh (see Figure 5.2b) was used for all the calculations. The different models corresponding to each interface shape/position were built from this mesh
by changing the assigned material properties (i.e. the electrical conductivity) of the elements in the mesh to create regions of solid, liquid or air.

In order to account for the skin effect at high frequencies, the finite element mesh was generated with an increased number of elements concentrated towards the edge of the charge. As a result, the elements with the smallest depth (0.038mm) were placed along the outer surface of the sample. These element sizes were smaller than the skin depth at the highest frequency analyzed (2MHz) for the most conductive sample condition (liquid Ge).

Figure 5.2 a) Finite element model geometry. b) Finite element mesh in interface region.
The applied load for this problem was the driving current in the multiple turn primary coil. This was specified as a point current at each of the 7 grid points corresponding to the location of each of the 7 turns on the primary coil. Since each calculation was normalized with respect to the empty coil condition, the actual value of current in the primary coil was not important and for convenience was taken to be unity. The model had a total of 913 grid points and 1007 (triangular and quadrilateral) elements. The output of the model allowed calculation of the inductive reactance of the coil. The model did not incorporate the capacitive reactance or the a.c. resistance of the coils, nor the impedance contributions of other test circuit elements.

The commercial axisymmetric finite element code solved equation (4.14) for the magnetic vector potential \( A(r, z) \), where \( r \) is the radial and \( z \) the axial position. The magnetic vector potential obtained from the finite element calculations was directly used to obtain the sensor's transfer impedance.

For an “absolute” sensor, equations (4.21) and (4.22) can be combined to yield,

\[
Z = \frac{4\pi^2 fN_s r_s}{I_p} [\text{Im}(A_{\text{ave}}) - j\text{Re}(A_{\text{ave}})]
\]  

(5.2)

where, \( r_s \) is the secondary coil radius, \( N_s \) is the number of turns in the “absolute” secondary coil, \( f \) is the test frequency (in hertz) and \( A_{\text{ave}} \) is the average vector potential over the cross section of the secondary coil wire.

For an axially separated “differential” sensor, the impedance is calculated from the algebraic sum of the two differential coil impedances.

\[
Z = \frac{4\pi^2 f r_s}{I_p} \left\{ N_s_1 [\text{Im}(A_{\text{ave}}) - j\text{Re}(A_{\text{ave}})]_1 - N_s_2 [\text{Im}(A_{\text{ave}}) - j\text{Re}(A_{\text{ave}})]_2 \right\}
\]  

(5.3)

where, \( N_s_1 \) and \( N_s_2 \) are the number of turns at the two locations of the differential secondary. All of the calculated coil impedances were normalized with respect to the coil impedance at the calculation frequency. This was obtained by replacing the relevant electromagnetic properties (\( \mu \) and \( \sigma \)) of the “solid” and “liquid” region elements of the
charge by those of the "air" elements, and repeating the finite element analysis.

5.4 Simulation results

5.4.1 Absolute sensor

5.4.1.1 Homogeneous liquid or solid states.

The simplest problems to analyze are the initial and final states of a growth run when the sensor observes either only the melt (prior to solidification) or only the solid (after completion of growth). In this case, the test material was assumed to have a uniform conductivity as defined in Table 5.1. Figure 5.3 shows the absolute sensor's calculated normalized impedance response for the liquid (circles) and solid (squares) states of CdTe, GaAs, Si and Ge at 13 frequencies between 200Hz and 2MHz. The impedance data for both the liquid and solid states of all four materials falls on the same characteristic "comma shaped" curve. The shape is almost identical to that expected for an infinite conducting cylinder contained in a long solenoid. The "size" of this curve can be characterized by its high frequency intercept (I) with the normalized imaginary impedance component axis. This is a function of the sample and pickup coil diameters, and is independent of the test material conductivity. For an infinitely long cylinder contained in a long solenoid, fringe field effects are insignificant and $I = 1 - \left(\frac{d_s}{d_c}\right)^2$ where $d_s$ is the sample diameter and $d_c$ the diameter of the secondary coil. Both the liquid and solid forms of all four test materials intercept the imaginary axis at the same point because the intercept is independent of the test material's conductivity in the high frequency limit. (In this limit all conductors totally exclude the penetration of flux into the sample.) This well understood phenomena is the basis for eddy current dimensional sensing and could be exploited in vertical Bridgman growth (e.g. to detect debonding of the solid from its ampoule during cooling).
Figure 5.3 Calculated absolute sensor impedance curves for the liquid and solid states of a) CdTe  b) GaAs  c) Si  d) Ge.

The only difference between the sensor's response to either a solid or liquid test material is a shifting of frequency points along the impedance curve. A decrease in conductivity, associated for example with solidification, causes the impedance at a fixed frequency to move counter clockwise around the curve because the sample becomes less inductive. This also explains why the length of the curves calculated up to 2MHz decrease as the test material conductivity decreases. In the limit, as the conductivity of the test material approaches zero, the normalized impedance at even the highest frequencies
would be located at \((0+j)\), i.e. at the upper left corner of the impedance plane, which is the same as the no sample ("coil in air") situation.

The imaginary component of impedance (i.e. the normalized inductive reactance) is plotted as a function of frequency for the liquid and solid states of the four test materials in Figure 5.4. At low frequencies (e.g. below 10kHz for CdTe), each material system gives a null response. This arises because the rate of change of the electromagnetic field within the test material is insufficient to induce detectable eddy currents. The sample is effectively transparent to the field, and the sensor's response is similar to that when no sample is present. Figure 5.5 shows vector potential contours for liquid CdTe for three test frequencies. Note that at a frequency of 10kHz, Figure 5.5(a), the vector potential contours are indistinguishable from those of an empty coil.

Figure 5.4 shows that beyond this threshold frequency, a clear separation of the liquid and solid impedance curves is seen, and a measurement of the imaginary impedance component in this region could be used to distinguish between the solid and liquid states. Beyond the threshold frequency, the separation of the curves is seen to at first increase, reach a maximum, and to finally decrease as the frequency is increased. For the highest conductivity material (Ge), the liquid/solid impedance separation decreases more rapidly as the test material conductivity increases because the skin effect more effectively expels flux in higher conductivity materials. This flux expulsion can be clearly seen in the vector potential contour plots of Figures 5.5(b) and 5.5(c). The impedance of the sensor in the intermediate range of frequencies (where detectable eddy currents are excited in the sample but flux expulsion is not complete) depends both on the test sample's diameter and its conductivity. Data collected at these frequencies (where skin depths are around 0.5 times the sample radius) is widely used to measure the conductivity of test materials of known diameter (obtained from high frequency data) and to infer sample conditions that affect it (e.g. temperature)[161,162].
Figure 5.4 Imaginary component of impedance vs. frequency for the liquid and solid states. a) CdTe b) GaAs c) Si d) Ge.

The solid and liquid impedance curves represent the sensor's response to two extremes of a growth process; they are the "upper" and "lower" bounds of all sensor responses that could be encountered during growth. All the impedance curves observed, no matter what interface shape or position, must lie between these bounds. The relative sensitivity of an eddy current solidification sensor to interfacial shape/position will depend upon the magnitude of separation of the liquid and solid impedance curves (ImΔZ) which is a function of the excitation frequency. Examination of Figure 5.4 reveals that
there exists a characteristic frequency where a maximum impedance separation (and thus, sensor sensitivity) occurs. Values for $Im \Delta Z_{max}$ and the frequency at which it occurs are tabulated for each of the test materials in Table 5.2.

![Figure 5.5 Magnetic Vector Potential contours for liquid CdTe at frequencies of, a) 10kHz b) 500kHz c) 2MHz.](image)

Table 5.2: The maximum difference between the imaginary impedance components ($Im \Delta Z_{max}$) of an absolute sensor for the homogeneous liquid and solid states and the frequency at which it occurs.

<table>
<thead>
<tr>
<th>Material</th>
<th>CdTe</th>
<th>GaAs</th>
<th>Si</th>
<th>Ge</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Im \Delta Z_{max}$</td>
<td>0.2417</td>
<td>0.3817</td>
<td>0.3625</td>
<td>0.3167</td>
</tr>
<tr>
<td>Frequency (Hz)</td>
<td>$2.5 \times 10^5$</td>
<td>$7.5 \times 10^3$</td>
<td>$4.5 \times 10^3$</td>
<td>$2.2 \times 10^3$</td>
</tr>
</tbody>
</table>

Examination of Tables 5.1 and 5.2 reveals that $Im \Delta Z_{max}$ monotonically increases with the liquid to solid conductivity ratio, Figure 5.6. The frequency at which the maximum differences occur varies inversely with the melt (or solid) conductivity. Germanium (which has the highest liquid and solid conductivities) has the lowest frequency where the maximum imaginary impedance change occurs. The highest
frequency occurs in CdTe which has the lowest liquid and solid conductivities. Clearly, the best frequency for operation of an eddy current solidification sensor is a material dependent parameter, and varies from one system to another. All of the four systems analyzed here have a sufficiently high conductivity that the frequency of “maximum response” is well within the range of frequencies experimentally accessible with conventional eddy current sensing instrumentation and have a sufficiently large $\text{Im} \Delta Z_{\text{max}}$ value for reliable eddy current sensing.

Figure 5.6 The variation of the imaginary component of impedance with the liquid/solid conductivity ratio for the absolute sensor.
5.4.1.2 Interface position effects.

To assess the response of an absolute sensor to the position of an interface, a series of calculations were performed for five locations of a flat interface. Figure 5.7 shows calculated normalized impedance curves for three of these positions. The impedances of all four materials are seen to converge at high frequency and again approach a common intercept with the imaginary axis (because in the modelled problem all four materials have the same diameter and therefore identical fill factors). In the limit, as the test frequency approaches infinity, the sensor's response depends only upon this fill factor and is independent of the interface's position within it, even when a liquid-solid interface exists within the interrogated volume.

The impedance curves are seen to be a strong function of interface position at lower frequencies, Figure 5.7. Recall that in the completely liquid or solid cases (Figure 5.3), the sample acted like an infinite cylinder of uniform conductivity encircled by a long solenoid, and changes of conductivity only shifted the impedances at specific frequencies around a common curve. However, when an interface between dissimilar conductivity materials exists within the field of an eddy current sensor, the observed response can be viewed as the net effect of simultaneous interactions with two finite length cylinders of different conductivities. Fringing of the field at the interface allows the impedance for a given frequency to move at a non-zero angle to the characteristic impedance curve, in effect shrinking its size. The contribution of this effect must depend upon where the interface is located with respect to the sensor. Thus, when the interface is below the center point of the sensor (say, h=−12.7mm, Figure 5.7), more of the higher conductivity "liquid" cylinder is sampled by the encircling eddy current sensor. The "solid" cylinder is still in the field of view of the sensor, but contributes less to the sensor's response. Later in a growth process when the interface has grown upward beyond the center of the (stationary) sensor, say to a position h=12.7mm above the sensor's center, more of the lower conductivity "solid" cylinder is encircled by the sensor and a lesser contribution is made by the liquid region.
Figure 5.7 Normalized impedance curves for three positions of a flat interface for the absolute sensor, a) CdTe  b) GaAs  c) Si  d) Ge.

The consequence of this phenomenon can be clearly seen in magnetic vector potential contours. Figure 5.8 shows the 500kHz vector potential field for the CdTe material system for interface heights of -12.7, 0 and +12.7mm. Because of the bigger skin depth in the solid phase, the depth of penetration into the solid is always much greater than that of the liquid. Since the vector potential is continuous across the liquid/solid interface, the field near the interface is perturbed from that expected for a homogeneous cylinder of either conductivity. The extent of this perturbation depends on the frequency of excitation
(through the skin effect) and the relative position of the interface within the sensing coil. Since the fields are no longer the same as those of an infinite uniform cylinder, the sensor's response departs from that of an "ideal" uniform cylinder (Figure 5.3) and provides the potential for a method of sensing position.

Figure 5.8 Magnetic Vector Potential contours for CdTe at a frequency of 500kHz for a flat interface position at a) h=-12.7mm b) h=0mm c) h=+12.7mm.

Since this behavior again originates from the skin effect, the frequency at which it occurs will be conductivity (and thus test material) dependent. This can be seen more clearly by plotting the normalized imaginary impedance component against excitation frequency for each interface position, Figure 5.9. Again, there exist material dependent low and high frequency thresholds below/above which the sensor's response is independent of frequency. However, for each material there also exists an intermediate range of frequencies where sensitivity to interfacial position is a maximum. The sensitivity (i.e. the difference in impedance for h=±12.7mm) and optimal frequency are listed in Table 5.3 for each test material. For the highest conductivity materials (like germanium or silicon), the curves converge at significantly lower frequencies than for lower conductivity materials such as CdTe. This is because the penetration depth of the electromagnetic field
becomes infinitesimal (i.e. approaches the infinite frequency limit) at lower frequencies in higher conductivity materials. From Tables 5.1 and 5.3 it is also observed that the maximum separation due to interfacial position \((\text{Im} \Delta Z_{\text{max}})\), increases/decreases as the \(\sigma_{\text{liquid}}/\sigma_{\text{solid}}\) ratio increases/decreases. The frequency at which this maximum position effect occurs varies inversely with either the liquid or solid electrical conductivity.

Figure 5.9 Variation of the imaginary component of impedance with frequency for an absolute sensor for five positions of the flat interface. a) CdTe  b) GaAs  c) Si  d) Ge.
Table 5.3: Imaginary impedance component values at the frequency of maximum sensitivity to interface position for the absolute sensor.

<table>
<thead>
<tr>
<th>Position of the interface, h (mm)</th>
<th>CdTe</th>
<th>GaAs</th>
<th>Si</th>
<th>Ge</th>
</tr>
</thead>
<tbody>
<tr>
<td>-12.7</td>
<td>0.610</td>
<td>0.567</td>
<td>0.570</td>
<td>0.598</td>
</tr>
<tr>
<td>-6.4</td>
<td>0.640</td>
<td>0.600</td>
<td>0.602</td>
<td>0.628</td>
</tr>
<tr>
<td>0</td>
<td>0.682</td>
<td>0.654</td>
<td>0.654</td>
<td>0.674</td>
</tr>
<tr>
<td>+6.4</td>
<td>0.734</td>
<td>0.722</td>
<td>0.717</td>
<td>0.728</td>
</tr>
<tr>
<td>+12.7</td>
<td>0.778</td>
<td>0.782</td>
<td>0.774</td>
<td>0.773</td>
</tr>
<tr>
<td>ImΔZ_{max}</td>
<td>0.168</td>
<td>0.215</td>
<td>0.204</td>
<td>0.175</td>
</tr>
<tr>
<td>Frequency (Hz)</td>
<td>4x10^5</td>
<td>1x10^4</td>
<td>6x10^3</td>
<td>3.4x10^3</td>
</tr>
</tbody>
</table>

Figure 5.9 shows that in the intermediate range of frequencies, the sensor’s imaginary impedance component is a monotonic function of interface position. If such a sensor were used to monitor an interface that moved through the sensor, the “sampled” fractions of liquid and solid would keep changing within the volume interrogated by the sensor. When the interface was well below the sensor (say h=-12.7mm), a larger fraction of the high conductivity liquid would be sampled (in the limit of h tending to minus infinity, a uniform liquid response like that of Figure 5.4 would be obtained). As the interface approached the sensor, a progressively increasing fraction of solid would be “sensed”, and as the interface moved past the sensor, its response would approach that for a uniform solid, Figure 5.4. Since the liquid conductivity is always much greater than the solid’s, the net effect would always be an increase in the imaginary impedance as each of the liquids studied gradually turned into solid during the growth process. The differences in the imaginary impedance component when solidification occurs (Table 5.3) are well within the measurement sensitivity of eddy current instrumentation.
5.4.1.3 Interface shape effects.

During Bridgman growth the liquid-solid interface shape can be concave, flat or convex and can change curvature as the governing heat and fluid flow conditions evolve during growth [44]. In order to assess the response of the absolute sensor to this curvature, a series of calculations have been conducted where the interface shape has been allowed to have one of five shapes specified by a convexity parameter, \( \theta = z/D \), where \( z \) is the difference in axial intersection of the interface central axis and periphery of the sample (i.e. the interface height), and \( D \) is the diameter of the sample (see Figure 5.2). For each calculation, the point where the interface intersected the outer boundary of the test material was fixed at the axial location of the secondary coil (i.e., \( h=0 \)).

Figure 5.10 shows examples of the normalized impedance curves for a convex \( (\theta = +0.333) \), a flat \( (\theta = 0.0) \), and a concave \( (\theta = -0.333) \) interface. The shape of the interface is seen to have a small but significant effect upon the structure of the impedance plane curve. The dependence upon interfacial curvature disappears at low and high frequencies and exhibits a similar frequency dependence to the interface position effect. This can be more clearly seen when the imaginary impedance component is plotted as a function of frequency for the five interface shapes, Figure 5.11.

The dependence of the intermediate frequency impedance upon interface curvature again results from the electromagnetic flux interaction with each interface, Figure 5.12. In the infinite frequency limit, the flux is fully expelled from the sample, and the sensor's response is insensitive to the internal interface shape. This limit is most nearly approached in the higher conductivity materials (Ge, Si, GaAs) for frequencies beyond 1MHz. For lower conductivity materials such as CdTe, it would be necessary to increase the frequency toward 10MHz in order to obtain an impedance that is almost independent of interface shape.
Figure 5.10 Normalized impedance curves for three interface shapes for the absolute sensor. a) CdTe  b) GaAs  c) Si  d) Ge.

At lower frequencies, the sensor's imaginary impedance shows a significant dependence upon interfacial curvature. The frequencies at which the interface shape effect is a maximum and the magnitude of the impedance changes are both given in Table 5.4 for each of the test materials.
Figure 5.11 Variation of imaginary component of impedance with frequency for five interface shapes for the absolute sensor. a) CdTe  b) GaAs  c) Si  d) Ge.
Although the effect of curvature upon the imaginary impedance-frequency relationship was similar to that seen for interface location, the maximum interface shape effects occurred at a lower frequency than the interface position effect for all four materials. To understand why this occurs, recall that all of the interfaces meet at the same point on the test sample's outer boundary. It is only when the magnetic flux penetrates sufficiently deep into the material that it samples the interior solid-liquid boundary that each interface will differently perturb the flux at the secondary coil location. In CdTe, this is seen to occur at ~500KHz, Figure 5.12. In contrast, the interface position still affects the response of the sensor even when the flux is concentrated very close to the edge of crystal, i.e. when operating at higher frequencies. It is only when the infinite frequency limit is approached (and the sample truly behaves as a perfect reflector) that one loses sensitivity to the position.

![Figure 5.12 Magnetic Vector Potential contours for CdTe at a frequency of 500kHz for three interface shapes. a) \( \theta = +0.333 \)  b) \( \theta = 0 \)  c) \( \theta = -0.333 \).]
Table 5.4: Imaginary impedance component values where maximum interface shape effect occurs for the absolute sensor.

<table>
<thead>
<tr>
<th>Interface convexity, ( \theta )</th>
<th>CdTe</th>
<th>GaAs</th>
<th>Si</th>
<th>Ge</th>
</tr>
</thead>
<tbody>
<tr>
<td>+0.333</td>
<td>0.818</td>
<td>0.808</td>
<td>0.818</td>
<td>0.824</td>
</tr>
<tr>
<td>+0.167</td>
<td>0.786</td>
<td>0.769</td>
<td>0.780</td>
<td>0.790</td>
</tr>
<tr>
<td>0</td>
<td>0.762</td>
<td>0.740</td>
<td>0.752</td>
<td>0.765</td>
</tr>
<tr>
<td>-0.167</td>
<td>0.738</td>
<td>0.714</td>
<td>0.726</td>
<td>0.740</td>
</tr>
<tr>
<td>-0.333</td>
<td>0.711</td>
<td>0.684</td>
<td>0.696</td>
<td>0.712</td>
</tr>
<tr>
<td>( \text{Im}\Delta Z_{\text{max}} )</td>
<td>0.107</td>
<td>0.124</td>
<td>0.122</td>
<td>0.112</td>
</tr>
<tr>
<td>Frequency (Hz)</td>
<td>(2 \times 10^5)</td>
<td>(3.7 \times 10^3)</td>
<td>(2.1 \times 10^3)</td>
<td>(1.5 \times 10^3)</td>
</tr>
</tbody>
</table>

This analysis of an absolute sensor's response has shown it to be sensitive to both the position and shape of the interface. The sensitivity to both phenomena is frequency dependent and a maximum sensitivity exists at intermediate frequencies. The analysis has shown that both location and position effects are coupled in an impedance measurement in the intermediate frequency range. However, careful measurements over a range of frequencies may be able to separately resolve the two growth parameters because of their different frequency dependencies.

5.4.2. Differential sensor

The essential idea of an axially displaced differential sensor is to sample the difference in field perturbation at two positions along the axis of a sample. By placing two opposingly wound secondary coils at these locations and ensuring that they are symmetrically located within the primary coil, equal magnitude, but opposite sign voltages are induced in the coils when a homogeneous sample is present. The introduction of an inhomogeneous sample with different conductivities near the two pickup coils will perturb the electromagnetic flux at one coil more than the other, and a non-zero resultant voltage will be observed. Thus, such a sensor will be incapable of distinguishing between an entirely liquid or solid sample (because of equal but opposite induced voltages at the
two coil locations), but might exhibit enhanced sensitivity to the location and curvature of
an interface separating materials of different electrical conductivity.

5.4.2.1 Interface position effects.

Figure 5.13 shows the effect upon the normalized impedance curve of moving a
flat interface through a differential sensor. For these calculations, the two secondary coils
were placed close to either end of the primary coil (they were 34mm apart). It can be seen
that the imaginary component of impedance at first increased with frequency, reached a
maximum and then decreased for each location. Figure 5.13 indicates that the frequency
corresponding to the maximum imaginary component was interface position dependent
(the frequency increased as the interface passed upwards through the sensor). This can be
seen more clearly in Figure 5.14 where the imaginary impedance component is plotted as
a function of frequency for each h-value. At or above frequency of maximum response, the
impedance reached its maximum value well after the interface had passed through the
center of the primary coil. The exact location at which this occurred was determined by the
relative magnetic vector potentials at each secondary coil location. This depends on the
test frequency and the electrical conductivities of the solid and liquid. As the frequency is
further increased beyond this peak, the curves for each interface location continue to
remain separated until very high frequencies, again due to differences in the fringe field at
the two coil locations.
Figure 5.13 Normalized impedance curves for three positions of a flat interface for the differential sensor. a) CdTe  b) GaAs  c) Si  d) Ge.
Figure 5.14 Variation of the imaginary component of impedance with frequency for a differential sensor for five positions of a flat interface. a) CdTe  b) GaAs  c) Si  d) Ge.
5.4.2.2 Interface shape effects

Interface shape effects were investigated by changing the interface shape while maintaining the outer edge of the interface midway along the primary coil. The normalized impedance curves for concave, flat and convex interfaces are shown in Figure 5.15. The size of the impedance curve was observed to increase as the interface curvature changed from concave to convex. The sensitivity to interface shape at first increased with frequency, went through a maximum at a material dependent frequency, and then decreased again at high frequency (beyond 2MHz) before the curves eventually converged, Figure 5.15(a). This can be seen more clearly in Figure 5.16 which shows the imaginary impedance component's frequency dependence. These calculations reveal the existence of a relatively narrow, material specific, intermediate range of frequencies where a strong sensitivity to the interface's curvature exists. In this region, the imaginary impedance component monotonically increases as the interface's shape changes from concave to convex. Above and below this region the sensor has little or no sensitivity to curvature.

If Figs. 5.14 and 5.16 are compared, it is again apparent that the calculated impedance above $10^5$Hz ($10^7$Hz for CdTe) is dominated by the interface's location while lower frequency data is sensitive to both the interface curvature and the position. Therefore, data collected over a range of frequencies may be sufficient to separately discriminate interface location and shape. The range of frequencies where the sensor significantly responds to interfacial curvature is seen to be reduced with the differential sensor arrangement because of fringe field effects at the ends of the primary coil. The magnitude of the imaginary impedance component's change associated either with movement of the interface or a change of its curvature are also significantly enhanced with the differential sensor design (compare the ordinate scales of Figs. 5.10 and 5.16). Thus, from a practical point of view, higher quality information about interfacial curvature and location might be obtained with a differential sensor. However, this method would preclude the measurement of conductivity and the things that affect it (e.g., melt composition or temperature).
Figure 5.15 Normalized impedance curves for three interface shapes for the differential sensor. a) CdTe  b) GaAs  c) Si  d) Ge.
Figure 5.16 Variation of imaginary component of impedance with frequency for five interface shapes for the differential sensor: a) CdTe  b) GaAs  c) Si  d) Ge.
5.5 Summary

Axisymmetric electromagnetic finite element analysis has been used to investigate eddy current sensing approaches for determining the liquid-solid interface position and shape. Two encircling sensor designs have been analyzed in detail. The best frequency ranges for interface position detection were observed to be consistently higher than those for interface shape detection for all the semiconducting materials studied. These frequencies were found to be dependent on the liquid and solid conductivities of each material system. They increased as the materials systems’ conductivity decreased. Because inductive contributions to an eddy current sensor’s test circuit eventually become overwhelmed by parasitics and other circuit component impedances at high frequencies, the eddy current sensing method appears to be most promising for high liquid conductivity materials like Ge, Si and GaAs. Lower conductivity systems such as CdTe would require careful test circuit design to enable observations at the high frequencies predicted to be needed for location determination. Axially separated differential coils are more sensitive to changes in the position and curvature of the liquid/solid than secondary coil (absolute) sensor design.

The limitations of the electromagnetic finite element model is the inability to account for the significance of other test circuit impedances which can make important contributions to a sensor’s response at high test frequencies and potentially mask the contributions of the interface. An experimental study which evaluates the concepts proposed by the modelling approach and which determines the significance of the test circuit’s impedance and parasitic contributions to sensor responses is presented in Chapter 6.
Chapter 6: Model System Experiments

6.1 Introduction

The modelling study in Chapter 5 revealed that the success of an eddy current approach depends on the liquid's electrical conductivity and the liquid:solid conductivity ratio. CdTe had the lowest liquid electrical conductivity of the materials evaluated, and was concluded to be the most difficult candidate for eddy current sensing. However, the study suggested that even for this material, the approach might be successful provided relatively high frequencies were used (e.g. 0.1-5MHz). The principal concern was the (unmodelled) significance of other test circuit impedances which can make important contributions to a sensor's response at high test frequencies and potentially mask the contributions of the interface. Thus, an experimental study was conducted to experimentally evaluate the concepts proposed by the modelling approach and to determine the significance of the test circuit's impedance and parasitic contributions to sensor responses obtained during the monitoring of a low electrical conductivity system such as CdTe.

Since the growth of high purity CdTe crystals by the vertical Bridgman method is costly, typically takes several days, and the interface position and shape are not independently known, it is expensive, time consuming, and problematic to try and validate a proposed sensor's response via crystal growth experiments. Instead, experiments have been conducted with a model system consisting of pairs of silicon samples that were carefully machined to create various interface geometries. By doping the silicon crystals it was possible to achieve electrical conductivities similar to those of either solid or liquid CdTe at its melting point and thus create an electromagnetically equivalent problem to the one encountered in the growth environment. This could be done while still retaining a complete description of the test system's geometry and all of its relevant physical properties. The sensor's response to this model test system was then calculated and compared to that of the experimental setup.
6.2 Experimental procedures.

6.2.1. Samples

The model test systems were built using four 76.2mm diameter, <111> direction oriented doped silicon samples with resistivities representative of either the liquid or solid phases of CdTe at its melting point[108]. The supplier's (Lattice Materials Corporation) specifications for the two low resistivity samples (representative of the liquid) were 0.01±0.005 ohm-cm while the two high resistivity (i.e. solid) samples had resistivities of 0.25±0.05 ohm-cm. These are equivalent to sample conductivities in the rather broad ranges of 6,667-20,000 S/m and 333-500 S/m respectively. Subsequent four point probe measurements indicated that the two lower conductivity samples had measured conductivities of 360-432 S/m while the two higher conductivity samples had conductivities of 9246-10227 S/m.

The samples were machined so that the ends of one pair of cylindrical samples (one of low conductivity and one of high conductivity) formed matching convex and concave surfaces. The other pair of samples had flat surfaces, Fig. 6.1. By pairing the samples shown in Figs. 6.1(a) and (b) it was possible to simulate a flat interface. By placing the sample in Fig. 6.1(d) on top of that in Fig. 6.1(c) it was possible to represent a convex solid-concave liquid interface and by reversing the order of samples in Figs. 6.1(c) and (d), a concave solid-convex liquid interface could be obtained. An interface curvature parameter, θ, was defined as the ratio of the interface height, z (=0 and ±25.4mm), to the crystal diameter, D (76.2mm). This collection of test samples allowed assessment of interfaces with θ = ± 0.33 and θ=0.
Figure 6.1 Cylindrical silicon samples used to construct test material geometries with electrical conductivity discontinuities analogous to those encountered in the vertical Bridgman growth of CdTe.

6.2.2. Sensor designs

Two sensor designs were constructed using the same primary coil for both sensors, Fig. 6.2. The 7 multiple turn primary coil was wound on a 101.6mm outer diameter x 95.25mm inner diameter x 50.8mm long PMMA preform. A 1.04mm wide, 0.508mm deep, 7 turn helical groove with a spacing of 4 turns per inch was machined on the outer preform surface to secure the primary coil windings and prevent movement of the individual turns. Seven turns of 18 gauge (1mm diameter) copper wire were wound on the machined preform to give a primary coil of 101.6mm diameter.
Figure 6.2 The geometry of the model problem. An axisymmetric seven turn primary coil in conjunction with either a central coaxial pickup coil (absolute sensor) or a pair of opposingly wound coaxial pickup coils (differential sensor) interrogates a cylinder with regions of low (representative of solidified CdTe) and high (representing liquid CdTe) conductivity.

The secondary coil preform had a 95.25mm outer diameter, a 88.9mm inner diameter and a 50.8mm length. The middle section of its outer surface was reduced to 91.2mm to provide an annular gap for the placement of both the absolute and differential pickup coil wires. For the absolute sensor measurements, a 0.5mm diameter (24 gauge) single loop of copper wire was wound level with the center turn of the primary coil. For the differential sensor, a single turn of 24 gauge wire was wound level with the first turn of the primary coil in a clockwise direction. One end of the wire was then taken along the inner surface of the secondary coil preform to a location halfway between the sixth and seventh primary coil turns and wound in the counterclockwise direction. For some tests, the position of the lower turn was systematically varied to assess the significance of relative coil position on the sensor's performance.
6.2.3. Sensor instrumentation

The multifrequency transfer impedance of both sensor designs has been measured by assembling an eddy current instrument from off the shelf circuit testing components, Fig. 6.3(a). A model HP4194A multifrequency Impedance/Gain-Phase analyzer operating in the gain-phase mode was used to make gain and phase measurements. The primary solenoid was driven by the impedance analyzer's oscillator to impress an alternating electromagnetic field on the test material over a wide range of frequencies (50kHz-10MHz). A model 25A100 Amplifier Research Inc. RF power amplifier was used to enhance the drive signal strength at low frequencies. The current flowing through the primary coil was obtained by recording the voltage drop across a low inductance precision (1 ohm) resistor connected in series with the primary coil. This measurement was recorded on the reference channel of the analyzer. The emf induced in the secondary coil was measured on the analyzer's test channel. The analyzer was programmed to record the amplitude ratios of the test and reference channel voltages, and to then compute their gain, g, and their phase angle difference, φ, for 101 logarithmically spaced frequencies starting at 50kHz and ending at 10MHz. A total of 32 samples were averaged with an integration time of 5ms for each sweep frequency.

A complete set of gain/phase measurements were obtained first without a test sample (to give a reference empty coil reading for the gain, g₀, and phase, φ₀, at each frequency). The test sample was then inserted and the gain and phase re-measured. For each test frequency, the real (Re) and imaginary (Im) normalized impedance (Z) components were found from,

\[
Re(Z) = \left(\frac{g}{g_0}\right) \sin(\phi - \phi_0) \quad (6.1)
\]

\[
Im(Z) = \left(\frac{g}{g_0}\right) \cos(\phi - \phi_0) \quad (6.2)
\]
Figure 6.3 The eddy current test setup used a HP 4194A impedance analyzer to measure the multifrequency gain/phase response of the sensor. Data was normalized by the empty coil’s response and the resulting complex transfer impedance plotted on an impedance plane diagram.
The real and imaginary components of the transfer impedance (computed from equations 6.1 and 6.2) are functions of four independent measurements; g, the ratio (i.e. the gain), and \( \phi \), the phase difference of the test and reference channel voltages measured with a sample present, and \( g_0 \) and \( \phi_0 \), measured with the empty sensor. Each of these measurements has an associated measurement accuracy which depends on the input signal levels of the reference and test channels, the test frequency range, the number of samples averaged, and the integration time of the measurement [164]. The gain and phase measurement accuracies (\( \delta g \) and \( \delta \phi \)) can be computed from the reference and test channel voltage and phase accuracies specified in the HP4194A operation manual for a 1Mohm input impedance measurement[164]. According to the impedance/gain-phase analyzer manual, the measurement error could be improved if a 50ohm input impedance measurement configuration was used. However, this is obtained only at the expense of introducing a finite current in the secondary coil circuit of the sensor. At high temperatures, this would result in a sensitivity to temperature induced resistance changes in the secondary coil adversely affecting the measured test voltages. In addition, a lower input impedance would increase the "loading" effect of circuit components (resistance, capacitance, and inductance of the cables) between the pickup (secondary) coil(s) and the instrument.

Errors in both the empty and sample filled sensor gain and phase are incurred. If these errors are assumed to be random, the error in the imaginary impedance component is [165],

\[
\delta \text{Im}(Z) = \sqrt{\left( \frac{\partial}{\partial g} \text{Im}(Z) \right)^2 + \left( \frac{\partial}{\partial g_0} \text{Im}(Z) \right)^2 + \left( \frac{\partial}{\partial \phi} \text{Im}(Z) \right)^2 + \left( \frac{\partial}{\partial \phi_0} \text{Im}(Z) \right)^2}
\] (6.3)

The total error in the imaginary impedance component computed from these four measurements can be calculated by substituting equation 6.2 into equation 6.3. A similar expression can be used to compute the measurement uncertainty for the real impedance component by replacing the \( \text{Im}(Z) \) terms in equation 6.3 with \( \text{Re}(Z) \) and substituting equation 6.1 for \( \text{Re}(Z) \) in equation 6.3.
6.2.4. Experiments

An initial gain/phase measurement was made with the empty sensor mounted with its axis vertical. The effects of an interface's location were then investigated by placing one of the pairs of silicon samples (e.g. Figs. 6.1(a) and (b)) co-axially within the sensor and adjusting its height using a rack and pinion mechanism. This enabled systematic variation of the position of the interface relative to the center of the primary coil without adjustment of the sensor's position. It was a preferred test method since changing the sensor's position relative to a stationary sample might have introduced varying electromagnetic coupling between the test circuit lead wires which might have affected the self-inductance of the test circuit.

For the first measurements, the platform height was adjusted so that the outer edge of the surface of contact between the two cylinders was 12.7±0.025mm below the center of the primary coil. Gain and phase measurements were then made at this position. Keeping the sensor undisturbed, the height of the sample was subsequently increased in 6.35±0.025mm increments to a series of new positions and the gain/phase measurements repeated. In all, measurements were made for two positions of the interface below the sensor, one at the same level as the sensor's midpoint, and for two positions above the sensor.

To obtain insight into the significance of test circuit contributions to the sensor responses, the initial series of experiments were performed using 0.61m long connecting cables between the eddy current sensor and the gain/phase analyzer. The effect of the test circuit's capacitance, inductance and resistance was investigated by substituting longer coaxial cables (1.83m in length) and repeating the measurements.
6.3 Results

6.3.1 Absolute sensor

6.3.1.1 Interface position effects

The effect of interface location upon an absolute sensor's response to a flat interface (θ=0.0) is summarized in Fig. 6.4. It shows experimentally measured normalized imaginary impedance versus frequency data (shown at every 5th frequency measurement) from the absolute sensor design when a flat interface was positioned at five different heights within the sensor. FEM predicted responses using the methodology described in Chapter 5 and the conductivities measured with the four probe technique are also shown for comparison (as solid lines). The position of the interface can be characterized by a distance, h, measured from the center of the (stationary) primary coil to the interface, Fig. 6.2.

The five curves span a total interface translation of 25.4mm. The curve corresponding to h=-12.7mm might represent a situation encountered early during crystal growth before the interface had advanced significantly into the sensor. At this stage, the electromagnetic field of the sensor samples more of the higher conductivity liquid resulting in a large inductance change and the biggest shift (from unity) in the imaginary impedance for all of the test frequencies. An upward shift in the imaginary impedance component as the interface translated upwards through the sensor was seen for all test frequencies. This is consistent with a reduction in the “effective” conductivity sampled by the sensor’s electromagnetic field as the lower conductivity material propagates through the sensor. A maximum sensor sensitivity to position was observed at a frequency around 600kHz. Above this frequency, the curves for each position converged slowly. Even at 5MHz, a significant dependence upon location was seen. Below 600kHz, the curves for the different locations converged rapidly and showed no dependence upon location below about 10kHz.
Figure 6.4 A comparison of measured and FEM predicted imaginary impedance component frequency response for a flat interface located at various axial positions using an absolute sensor.

Excellent agreement between the model predictions and the experimental data was observed in the 5kHz-3MHz frequency range. Above 3MHz, the measured data deviated slightly from the calculated values and above 5.5MHz, experimental impedance values were observed to increase with frequency whereas the predicted data continued to decrease.

Table 6.1 shows the estimated uncertainties in the imaginary impedance component for the absolute sensor in the 5kHz-5MHz frequency range for θ=0.0 and h=0. The estimated error decreases from 0.032 at 5kHz to 0.018 at 5MHz and is approximately 3.25% of the nominal impedance at each frequency. For an operating frequency of around 700 kHz, the error analysis indicates an ability to sense a change in the position of a CdTe liquid-solid interface to within 2-3mm.
Table 6.1: Uncertainties in the imaginary impedance of the absolute sensor for $\theta=0.0$ and $h=0$

<table>
<thead>
<tr>
<th>Frequency (kHz)</th>
<th>Nominal Impedance</th>
<th>Probable Error</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>1.0003</td>
<td>0.0325</td>
</tr>
<tr>
<td>22.9</td>
<td>0.9836</td>
<td>0.0321</td>
</tr>
<tr>
<td>104.5</td>
<td>0.8675</td>
<td>0.0287</td>
</tr>
<tr>
<td>478.2</td>
<td>0.7203</td>
<td>0.0238</td>
</tr>
<tr>
<td>2186.7</td>
<td>0.6027</td>
<td>0.0199</td>
</tr>
<tr>
<td>5045.5</td>
<td>0.5435</td>
<td>0.0177</td>
</tr>
</tbody>
</table>

To understand the origin of the high frequency anomalous behavior of the experimental impedance curve, the gain and phase frequency responses for the empty sensor are shown in Fig. 6.5. The gain is seen to linearly vary with frequency up to about 3MHz as expected, but a non-linear behavior is observed at higher frequencies. A maximum gain occurred at 5.5MHz, after which a drop-off in the response was observed. An analogous effect in the phase response was also seen at high frequencies.

These anomalous responses are consistent with a circuit approaching resonance. The resonant frequency will be governed by the sensor's and circuit's lumped inductance, capacitance and resistance parameters. For the sensor, these depend upon the diameter of the sensor windings, the number of coil turns, the spacing between the windings, and the capacitive coupling between the two coils. Contributions from the lead wires (resistance, capacitance and inductance) connecting the sensor to the analyzer are length dependent. Thus in addition to the inductive coupling of the coils with the sample (which is calculated with the finite element method), frequency dependent parasitic capacitive effects, stray inductive effects, and lead wire impedance all contribute to the overall response as the frequency is increased.
Figure 6.5 The frequency response of the empty absolute sensor's gain and phase for different lengths of cabling connecting the sensor to its measurement instrumentation.

To confirm the origin of this anomalous behaviour, the coaxial cables were tripled in length (to 1.83m) and the empty sensor gain and phase measurements repeated. The transition to non-linear behavior was seen to occur at a slightly lower frequency when the length was increased. The magnitude of the deviation from linearity also increased as the lead length was increased. In the lower frequency region where the gain remained linear, very good agreement between the experimental results and the electromagnetic (finite element) model calculations was again seen. Fig. 6.6 shows a comparison of the electromagnetic FEM calculated impedance with experimental data obtained with both cable lengths (0.61 and 1.83m). It shows that significant discrepancies between model predictions and experimental measurements are likely to be encountered above 2MHz. However, even above 2MHz the trends associated with interface location are still evident, and so this effect does not necessarily preclude the use of higher frequency measurements,
but rather implies the need for careful calibration if the high frequency data is to be used to infer information about interface location.

![Graph showing the frequency dependence of an absolute sensor's imaginary impedance component for two test circuit cable lengths.](image)

**Figure 6.6** The frequency dependence of an absolute sensor's imaginary impedance component for two test circuit cable lengths.

These discrepancies between FEM predictions and the experiments are important only at higher frequencies (>2-3MHz). Earlier work in Chapter 5 showed that as the liquid conductivity decreases, it becomes increasingly necessary to perform measurements at higher frequencies to separate the contributions to the sensor's response from the interface's location from its shape (curvature). From the measurements reported above, the effective upper frequency that can be used without resorting to calibration is seen to be limited by circuit resonances. For high conductivity materials like GaAs, the test circuit effects occur at frequencies well above those that would be used for interface characterization. However, for low electrical conductivity materials like CdTe it is important to design the sensor and its test circuit so that these resonances are made to
occur at as high a frequency as possible to avoid adversely impacting the fidelity of the sensor's response in the region where it is dominated by interface location.

6.3.1.2 Interface shape effects

The effects of interface shape upon the measured and predicted sensor response for the absolute sensor are shown in Fig. 6.7. Very good agreement between measurements and predictions is again seen at frequencies below about 3MHz. In the frequency region where the agreement between measurement and prediction is good, changing the interface from concave through flat to convex leads to an increase in impedance component values. This is a direct result of the decrease in the fraction of the high conductivity liquid sampled by the sensor's electromagnetic field. For samples of the conductivity tested here, the strongest sensitivity to shape occurs at intermediate frequencies (~200kHz). In this frequency region, the calculated and measured responses are in excellent agreement and test circuit contributions can be safely ignored.

![Graph](image)

Figure 6.7 The effect of interface shape upon the measured and calculated imaginary impedance component of the absolute sensor for h=0.
As the frequency was increased, both the calculated and measured impedances converged as a consequence of the skin effect which confines the electromagnetic field within the sample to a thin annular shell near the outer surface of the sample. In this interrogated volume all the interfaces have similar fractions of high conductivity material and so the response to the detailed internal shape of the interface is lost.

At high frequency (>2-3MHz) the experimental data again deviated from the predicted behavior because of the approach of the measurement to a coil and test circuit resonance. Again, the relative trends predicted by the finite element method are seen to be still preserved even at the high frequencies where test circuit impedances and parasitic effects are significant. These unmodelled phenomena introduce discrepancies between the measurements and calculations. However, the data still appears useful, even when the theory is no longer strictly valid, because the predicted convergence of the responses for interfaces of differing shape is still preserved in the experimental data.

6.3.2. Differential Sensor

The response of a differential eddy current sensor is dependent on the locations of the two secondary coils, and their relation to the primary coil turns. If the secondary coils are placed exactly symmetrical with respect to the primary coil, the vector potentials of the empty sensor (or one containing a uniform conductivity sample) at the two pickup coil locations would be the same. Equal but opposite sign voltages would be induced in the two coils, and a zero net response would be obtained. If one of the secondary coils is slightly displaced in the axial direction, a small non-zero differential measurement is obtained. Now, when a non-uniformly conducting sample (e.g. one containing regions of liquid and solid with different electrical conductivities) is introduced into such a sensor, the symmetry of the electromagnetic field will be lost because of greater flux penetration into (low conductivity) solid regions. This distorted field results in a difference in vector potential values at the two coil locations and a non-zero resultant signal, even when the coils are symmetrically positioned with respect to the primary. In our data reduction scheme, the measurements made with the sample present are divided by the empty

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sensor's imaginary impedance, which depending upon the precise secondary coil locations, is close to zero. If the lower pickup coil is positioned slightly closer to the coil center (in a region of higher flux), and the higher conductivity material is always arranged to be on top, the normalized imaginary impedance values range from unity upwards as the interface is translated vertically through the sensor. Both an enhanced sensitivity to axial non-uniformity in the sample's conductivity and a lowering of the frequency where the response ceases to be interface shape dependent has been predicted for this differential sensor design.

6.3.2.1 Interface position effects

A flat liquid-solid interface constructed using the two samples shown in Figs. 6.1(a) and (b), was translated through a stationary differential sensor whose secondary coils were wound so that one was at the same level as the top turn of the primary coil and the second was level with the midpoint of the bottom two primary coil turns (giving a secondary coil separation of 34.9±0.025mm). Fig. 6.8 shows the imaginary impedance component frequency responses at five locations of the interface and compares them with finite element calculations. At each interface location, both the measured and predicted imaginary impedance component at first increased with frequency, reached a maximum and finally decreased with frequency for all the interface positions.

The peak in the frequency dependence of the imaginary impedance component arises because at low frequencies, weak eddy currents are induced in both solid and liquid, and the vector potentials at each secondary coil location are similar and almost identical to those of the empty coil. At very high frequencies, skin effects in both solid and liquid almost totally expel flux from the sample, and the vector potential at both coil locations again becomes the same, giving a null differential response like that of an empty coil. At intermediate frequencies, the primary coil's fringe field penetration in material at the two coil locations is different. Therefore, the two pickup coils experience a different vector potential leading to a non-zero response, and a peak in the frequency response.
Figure 6.8 The effect of interface location on the measured and predicted frequency response of a differential sensor. (a) h = -12.7 mm. (b) h = -6.35 mm. (c) h = 0 mm. (d) h = +6.35 mm. (e) h = +12.7 mm
A comparison of Figs. 6.8(a) through 6.8(d) shows an increase in the magnitude of the imaginary impedance peak as the interface begins to propagate upwards through the sensor. When the interface is about 6mm above the sensor's center, Fig. 6.8(d), the imaginary impedance component reaches a maximum and then declines, Fig. 6.8(e). This phenomenon can be understood by again examining the changes in the differential voltage resulting from variations in the vector potential at each of the two pickup coil locations. Suppose we have a stationary sensor and an upward moving interface. At first, both secondary coils would be located adjacent to high conductivity liquid and nearly equal but opposite voltages would be induced in the two coils. As the interface approaches or moves beyond the lower secondary coil position, the voltage induced in the lower coil begins to be dominated by (reduced) eddy current induction in the nearby solid, while the top coil still "samples" the vector potential resulting from the eddy current distribution in the liquid. As the interface continues to move beyond the lower secondary coil, the lower coil's response is increasingly dominated by the solid region's response to the impressed field and the difference in the two secondary coils' induction increases. Eventually, an interface position is reached where the lower coil completely ceases to sense the liquid, and the upper coil begins to "see" increasing amounts of the solid. The vector potentials at the two locations then begin to converge, and the impedance starts decreasing. For the selected values of conductivities in these experiments, this effect results in a maximum sensor response near h=+6.4mm.

The errors associated with the differential sensor impedance measurements are shown in Table 6.2 for the case of a flat interface located at h=0.0mm. The estimated error first increases from 0.034 at 5kHz to 0.104 at 478kHz and then decreases to 0.058 at 5MHz and is again approximately 3.25% of the nominal impedance value.

All of the results shown in Fig. 6.8 exhibited a discrepancy between the experimental data and calculated curves at high frequencies. The measured gain/phase response of this differential sensor, Fig. 6.9, shows a non-linear gain-frequency response above 1MHz. Small deviations in phase are evident at even lower frequencies (~400kHz).
When converted to impedance, this results in smaller than predicted imaginary impedance component values at high test frequencies and arises because of the larger "parasitic" impedance contributions in a two coil system.

<table>
<thead>
<tr>
<th>Table 6.2: Uncertainties in the imaginary impedance of the differential sensor for $\theta=0.0$ and $h=0.0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Frequency (kHz)</td>
</tr>
<tr>
<td>-----------------</td>
</tr>
<tr>
<td>5</td>
</tr>
<tr>
<td>22.9</td>
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<tr>
<td>104.5</td>
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<tr>
<td>478.2</td>
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<tr>
<td>2186.7</td>
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<tr>
<td>5045.5</td>
</tr>
</tbody>
</table>

A comparison of the linear portions of the empty sensor gain curves for the absolute and differential sensors, Figs. 6.5 and 6.9, confirms that the voltages induced in the differential mode sensor (0.1-70mV) are smaller than those of the absolute sensor (1-300mV). The sensitivity of the differential sensor might be improved if the empty sensor's gain were further decreased (e.g. by positioning the two secondary coils in more symmetrical locations). Fig. 6.10(a) replots the experimental gain/phase data of Figs. 6.8(a)-(e) where the secondary coil spacing was 34.9mm. Figs. 6.10(b) and (c) show corresponding data for secondary coil spacings of 36.5$\pm$0.025mm and 33.3$\pm$0.025mm. The spacings were adjusted by moving the lower coil position by 1.6mm (1/16 in.) in either direction while keeping the position of the upper coil unchanged. A 36.5mm separation corresponds to the case of two coils that are very nearly symmetric with respect to the primary coil giving a very small "empty" differential voltage measurement. This results in smaller gain values over the entire frequency range (as shown by the dashed line with circles in Fig. 6.9), and the larger impedance values shown in Fig. 6.10(b). A
reduction in spacing (to 33.3±0.025mm) is seen to have the opposite effect, Fig. 6.10(c). The empty gain for this case (shown by the dashed line with "squared" symbols in Fig. 6.9) increased and the resulting impedance values decreased. The response of a differential sensor is clearly sensitive to the precise positioning of the secondary coils and this must be carefully controlled if the data is to be quantitatively compared with model predictions.

![Graph showing empty sensor gain and phase vs. frequency for three differential sensors with slightly different pickup coil spacings.]

Figure 6.9 The differential sensor's empty gain/phase frequency response for three differential sensors with slightly different pickup coil spacings.
Figure 6.10 A comparison of the responses of three differential sensors with slightly different pickup coil separations.

6.3.2.2 Interface shape effects

The interface shape effect (while keeping the position h=0) was experimentally investigated using a secondary coil spacing of 34.9mm. A comparison of the calculated and experimental results is shown in Fig. 6.11 for each interface shape. Since the interface position on the outer surface of the sample remained the same, the peaks in the impedance responses occurred around the same frequency (~600kHz). When the outer edge of the interface was positioned at the middle of the sensor (h=0.0mm), a large monotonic increase in the peak impedance value accompanied a change in convexity from -0.333 to +0.333.
Figure 6.11 Measured and predicted differential sensor responses for three different interface shapes.

Although exact agreement between the experimental data and the FEM calculation was not observed, the differences in the two sets of curves appear to be small. They may be due to a combination of random measurement error and a slight mispositioning of the two sensing coils during construction of the sensor. At high frequencies, the non-linear increase in gain (associated with the sensor resonance characteristics) again results in the experimental values that are consistently smaller than the calculations. Both the predicted impedance curves and the measured impedance data for each interface converged at high test frequency. In this regime, the sensor response is independent of the interface shape and depends only on interfacial location within the sensor. Test circuit effects shift the experiment's point of convergence downwards from the model’s predictions. Nonetheless, it still appears possible to operate this sensor in high frequency regions (~5MHz) where its
response is dominated by interface location and at lower frequencies (600kHz) where it senses both location and shape.

6.4 Summary

Benchtop eddy current measurements have been conducted with 76mm diameter doped silicon single crystals to explore the responses of absolute and differential sensors during vertical Bridgman growth of CdTe like semiconductors. They have been compared to finite element electromagnetic model calculations of the experimental test setup and very good agreement was obtained over a wide range of interface positions and interface shapes for both sensor designs. Both sensors have a maximum response to interface position and curvature at around 600kHz. At higher frequencies, the measurements were increasingly independent of interface shape and depended only upon location. However, at the highest test frequencies, unmodelled circuit effects significantly contribute to the sensor’s response and a departure between FEM predicted and measured behavior exists. In this region, it will be necessary to use either an experimental calibration or include a complete circuit analysis in an improved model to avoid incurring errors in eddy current sensor deduced interface locations. Alternative approaches for separately discriminating interface position and shape contributions to the sensed response are explored in Chapter 7.
Chapter 7: Interface Shape and Location discrimination

7.1 Introduction

The modelling and experimental studies in Chapters 5 and 6 indicated that provided contributions from interface location and shape can be separated, the signals from eddy current sensors appear well suited for monitoring the growth of CdTe. Several monitoring strategies could be pursued. For example, a sensor could be positioned at a fixed location with respect to the ampoule, and the passage of the interface along the ampoule monitored, or the sensor could be continuously repositioned along the ampoule to coincide with the interface, and the sensor’s position then continuously monitored to track interface location (and measure its velocity). In either case, an anomaly-free protocol is needed for separately discriminating the location of the interface within the sensor and determine its curvature.

Here, the finite element method is used to calculate the multifrequency output of eddy current sensors as a liquid-solid interface is propagated upwards through the sensor. This simulation of a stationary sensor’s response during a simulated growth run is repeated for three different interfaces (flat, concave, and convex) and performed for two material systems (GaAs and CdTe). The modelling studies in Chapter 5 showed GaAs to be an ideal material system for eddy current sensing because its high conductivity enables the use of low frequencies where test circuit impedance/parasitics are absent during measurements. CdTe appeared to be more a marginal material system and represents a critical test of the eddy current method. The sensor responses from these simulated growth runs reveal two possible strategies for separating the contributions of the interface’s location from its curvature. One uses the frequency dependence of the complex impedance; the second exploits the observation of a frequency dependent inflection point/peak in the imaginary impedance-interface location relationship for the absolute/differential sensor designs. The location of these points is found to provide good
discrimination between interface location and curvature even for the more problematic CdTe material system.

### 7.2 Analysis methodology

The problem analyzed is shown in Fig. 7.1. It consists of a cylindrical sample (of diameter, \(D=76.2\text{mm}\)) contained within an axisymmetric eddy current sensor. The sample has two regions with electrical conductivities of either the solid or liquid semiconductor at its melting point. These conductivity values were the same as those reported earlier (Table 5.1). The interface between the two conductivity regions was allowed to be flat, concave, or convex with a convexity parameter \(\theta = z/D = \pm 0.333\) and 0 (\(z\) is the maximum difference in axial coordinates of the interface across the sample). The sample’s magnetic permeability (\(\mu\)) was taken to be \(4\pi \times 10^{-7}\text{H/m}^{-1}\) which is that of free space.

![Figure 7.1 Finite element model geometry.](image)

Figure 7.1 Finite element model geometry.
The multifrequency responses of two types of sensor were analyzed. Both used a 7 turn driver (or primary) coil with a 6.35mm coil spacing through which was passed a fluctuating unit current. The resulting electromagnetic field was calculated using the same FEM procedures described in Chapter 5. An absolute (single pickup coil) sensor response was obtained from the electromagnetic field's vector potential at the pickup coil located midway along the length of the primary coil, Fig. 7.1. The differential (two axially displaced, opposingly wound pickup coils) sensor response was obtained from the difference in the vector potentials at each pickup coil's location (near either end of the primary coil), Fig. 7.1.

The finite element model was used to first calculate the 500kHz responses of the absolute and differential sensors for a homogeneous liquid sample with the electrical conductivity of CdTe. A flat liquid-solid interface was then introduced at a height $h = 38.1$ mm below the sensor's midpoint (see Fig. 7.1 for the definition of $h$) and the two sensors' responses at 500kHz again calculated. The interface was systematically advanced in 3.2mm increments upwards through, and eventually beyond, the sensor and the two 500KHz responses obtained. Finally, the response due to an entirely solid sample was obtained. This procedure was repeated for ten other excitation frequencies to cover the 10kHz to 10MHz range, and for interfaces with convexities of ±0.333. The electrical conductivities of the test system were then changed to the values of GaAs, and the entire procedure was repeated for 12 frequencies ranging from 200Hz and 1MHz. Finally, the sensors' impedances with the samples removed were calculated to normalize sensor responses.

### 7.3 Absolute sensor

Fig. 7.2(a) shows the normalized imaginary component of the absolute sensor's impedance for CdTe at a test frequency of 500kHz as a function of interface location for each interface shape. Results for GaAs at 10kHz are shown in Fig. 7.2(b). At these two frequencies, both materials exhibit similar behaviors. Initially, when the interface is
beyond the range of the sensor's electromagnetic field (say \( h = -80 \text{mm} \)), the imaginary impedance has a value that depends only upon the liquid's electrical conductivity and the fill factor of the coil. If the coil's fill factor is known, the value of the impedance at a fixed frequency moves downwards from its null (i.e. empty coil) value of \( 0 + j1 \) as the conductivity increases. This data could be used to determine the liquid's electrical conductivity (and to thus infer the factors affecting it) during applications of the sensor to a real growth process. As the interface propagates upwards through the sensor, \( h \) goes from negative to positive, the impedance increases, goes through an inflection and asymptotically approaches a constant value corresponding to that of the solid. The impedance in this latter case again depends only upon the test material's electrical conductivity and the coil's fill factor. If the latter remains constant (i.e. the test sample and secondary coil diameters do not change), an impedance measurement in this region could be used to infer the solid's electrical conductivity and gain insight into the factors that control it (e.g. the average temperature in the sensed volume).

![Figure 7.2](image)

**Figure 7.2** Calculated impedance variation of the absolute sensor with interface position for simulated growth runs of three interface shapes. (a) CdTe (500 kHz) (b) GaAs (10 kHz).
The results presented in Fig. 7.2 indicate that when the interface is located far from the sensor's center (e.g. \( h = \pm 40\text{mm} \) or more), the imaginary impedance is approximately independent of both its location and curvature. However, as the interface approaches the location of the pickup coil, a strong dependence upon both location and curvature is seen. For both material systems, the imaginary impedance associated with a sample containing a convex interface first begins to increase (towards the sensor's null value of unity on the imaginary axis) as the interface approaches the sensor's midpoint from below. If the location of the interface (i.e. \( h \)) were known independently, the imaginary impedance component at these test frequencies (10kHz for GaAs and 500kHz for CdTe) is directly related to the interface's convexity. For example, if the outside edge of the interface coincided with the pickup coil location (\( h=0.0\text{mm} \)), the imaginary impedance for GaAs would increase from 0.615 for a concave interface to 0.715 for a convex one. A smaller, but still significant shift would occur for CdTe. It can also be seen that the point of inflection shifts to the left as the interface's curvature changes from concave to convex. Therefore, either the value of the impedance or the position of the inflection point are interfacial curvature dependent.

The response to interface location depends upon test frequency for both materials. Fig. 7.3(a) shows the imaginary impedance-position relationship for CdTe with a flat interface at test frequencies of 50kHz, 100kHz, 500kHz, and 5MHz. Analogous results for GaAs at 500Hz, 2kHz, 10kHz, and 1MHz are shown in Fig. 7.3(b). At very low frequencies (below 50kHz for CdTe and 500Hz for GaAs), the rate of change of the electromagnetic flux within the test material is sufficiently low that weak eddy current induction occurs, and both samples are almost electromagnetically transparent. In these cases, the decrease in electrical conductivity associated with passage of the liquid-solid interface through the sensor results in a very small change in the nearly null response of the sensor. At high frequency (above 1MHz for GaAs and 5MHz for CdTe), the very high rate of change of flux induces intense eddy currents that are concentrated close to the sample surface (due to the skin effect). In this frequency range, the sensor's response depends strongly on the coil's fill factor (i.e. the sample diameter) and progressively less
on conductivity as the frequency increases. Thus, the sensor’s response is only moderately affected by the location. The largest changes in response are seen at intermediate frequencies where both the eddy current density and the volume within which it exists are both large.

![Graphs of imaginary component of impedance vs. interface position for CdTe and GaAs](image)

Figure 7.3 Variation of the imaginary impedance component of the absolute sensor with interface position for a flat interface at four frequencies. (a) CdTe (b) GaAs.

Fig. 7.4 shows high and intermediate frequency results for each interface shape. It can be seen that for GaAs, Fig. 7.4(b), a 1MHz measurement is independent of interface shape but is a unique, though weak, function of location, whereas the intermediate frequency (10kHz) impedance monotonically increases as either the interface changes from concave to convex, or as its position moves upwards through the sensor. Thus, measurements at the two frequencies shown in Fig. 7.4(b), in combination with the pre-calculated responses for different interface shapes, could be used to separately determine the interface location and shape. The frequencies required to do this for GaAs are below those where test circuit impedance/sensor parasitics are likely to perturb the responses.
Figure 7.4 Variation of the imaginary impedance component of the absolute sensor with interface position for three interface shapes at two frequencies. (a) CdTe (b) GaAs.

CdTe exhibits a less ideal response, Fig. 7.4(a). Even at 5MHz (where test circuit impedance/sensor parasitics begin to significantly contribute to experimental measurements), a small residual dependence upon interface shape is observed. This results in greater uncertainty in the interface’s location determination, and since this needs to be known before the interface can be characterized, it results in a reduced ability to characterize the interface’s curvature. This is further compounded by the smaller differences in intermediate frequency (500kHz) sensor response to each interface shape. Measurements at very high frequency (suitably corrected for test circuit/sensor parasitic shifts) might overcome this difficulty, and the precision of the interfacial curvature could be improved by developing an analysis for a range of test frequencies.

An alternative, potentially simpler approach is to examine the inflection point in the imaginary component’s position dependence. It is clear from Figs. 7.3 and 7.4 that this is a function of test frequency and interface shape. To investigate it in more detail, the imaginary impedance-position relationships for each interface shape, frequency, and
material were numerically differentiated with respect to interface position and the resulting peak location (and thus the data’s inflection point) determined. This inflection point position is plotted versus test frequency for CdTe in Fig. 7.5(a) and for GaAs in Fig. 7.5(b).

Figure 7.5 Relative position of the inflection point on the imaginary impedance component response of the absolute sensor vs. frequency for three interface shapes. (a) CdTe (b) GaAs.

Fig. 7.5 shows that at high frequencies the inflection point position becomes a progressively weaker function of the interface shape. The inflection point for data collected at 1 or 2MHz during the simulated growth of GaAs corresponds to an interface that is level with the pickup coil location. This result is valid within ±0.5mm for all three interface shapes. At lower frequencies, the results of Fig. 7.5(b) indicate that a large shift in the inflection point occurs as the interface’s curvature changes from convex to concave. For this material system, measurement of the high frequency inflection point (for example, by axially translating the sensor along the ampoule) could be used to position the sensor at the interface’s location, and lower frequency data (say 10kHz) could then be used in conjunction with Fig. 7.5(b) to infer the curvature. If the axial position of the sensor were
simultaneously monitored, the strategy would enable separate discrimination of interface location and curvature.

The lower electrical conductivity CdTe system, Fig. 7.5(a), exhibits a similar behavior to GaAs but at significantly higher frequency. For this system, even inflection point data collected at 10MHz exhibits a ±1.5mm position variability due to interfacial curvature. This degree of uncertainty may still be sufficiently precise for some applications (for example, ensuring that solidification occurs at an optimal location in the furnace), but would enable only qualitative insights into the interface's curvature to be obtained from lower frequency (say 500kHz) data. Efforts to make measurements at higher frequency to reduce the variation in the inflection point data would have to contend with test circuit contributions to the overall response. These are likely to dominate measurements in the 10-25MHz range for most experimental setups. Alternatively, extrapolating inflection point data collected in the 1-10MHz range out to ~50MHz would reduce uncertainty in location and might enable a more precise determination of curvature.

7.4 Differential sensor

The physical basis of the discrimination approaches proposed above lies in the expulsion of electromagnetic flux by the skin effect. At high frequencies, a uniformly distributed excitation field of the primary coil exists near its center when no sample is present. When a solidification interface resides within the sensor, the skin depths at high frequency are small in both the solid and the liquid, and the electromagnetic field samples only the outer surface of the test material. In this situation, the interface's location deep within the sample has little or no interaction with the excitation field, and the sensed response depends only upon the fraction of solid and liquid in a thin annular region at the sample's surface (i.e. it depends only upon the location of the interface within the sensor and not the radial dependence of the location). Unfortunately, the low conductivity (large
skin depth) of CdTe requires the use of too high a frequency for this to be easily accomplished.

If this limitation is to be overcome, some way is needed to increase the rate of decay of the excitation field into the sample. In earlier modelling and experimental work, the placement of pickup coils at the ends of the excitation coil was shown to result in a more rapid convergence of the impedance with frequency for the different shaped interfaces. This arose because the "fringe" field at the top and bottom of the sensor decays more rapidly with radial distance in the sample than predicted by a skin effect alone. This additional mechanism of radial field decay can be exploited in the differential sensor approach to better discriminate between interfacial location and curvature.

Fig. 7.6 shows the variation in the imaginary impedance of a differential sensor when concave, flat, or convex interfaces are propagated through the sensor. The result for CdTe corresponds to an excitation of 500kHz whilst that for GaAs was obtained at 10kHz. In both material systems, the passage of the interface through the sensor results in a peak in the impedance. When the interface lies outside the sensor (so that the sensed region is either solid or liquid), a near null response is obtained. As the interface passes close to the sensor's midpoint, a peak in impedance is observed, and at the two frequencies referred to above, the position of the peak is a significant function of the interface's curvature. The position within the sensor where the peak is seen also depends upon the test frequency, Fig. 7.7.

The position dependence of the impedance at high and intermediate frequencies is shown for three interfaces in both CdTe and GaAs in Fig. 7.8. It can be seen that for both materials the high frequency data is nearly independent of interface curvature when the interface is located near the center of the sensor. At lower frequency, the position of the impedance peak is a strong function of the interface's position and curvature.
Figure 7.6 Calculated impedance variation of the differential sensor with interface position for simulated growth runs of three interface shapes. (a) CdTe (500 kHz) (b) GaAs (10 kHz).

Figure 7.7 Variation of the imaginary impedance component of the differential sensor with interface position for a flat interface at four frequencies. (a) CdTe (b) GaAs.
Figure 7.8 Variation of the imaginary impedance component of the differential sensor with interface position for three interface shapes at two frequencies. (a) CdTe (b) GaAs.

Fig. 7.9 shows the position of the impedance peak as a function of frequency for each interface and both materials. The fringe field effect combined with the differential scheme results in a lowering of the frequency at which the peak positions converge for both materials. Thus, using the results shown in Fig. 7.9, data collected at 500kHz could be used to locate the interface’s position to better than ±0.5mm in GaAs. For CdTe, data collected in the 5-10MHz range would enable the location to be deduced to better than ±1mm. For both material systems, lower frequency data (say 500kHz for CdTe and 10kHz for GaAs) could be used in conjunction with the calculations of Fig. 7.9 to deduce the interface’s curvature. The precision of the curvature characterization will depend upon the accuracy of the location determination. It is estimated to be on the order of 0.05 of the convexity parameter (i.e. approximately 4mm for a 76mm diameter sample) assuming the location is determined to ±1mm.
Figure 7.9 Relative position of the peak in the imaginary impedance component response of the differential sensor vs. frequency for three interface shapes. (a) CdTe (b) GaAs.

7.5 Summary

An electromagnetic finite element method has been used to calculate the response of absolute and differential eddy current sensors during the simulated vertical Bridgman growth of CdTe and GaAs. GaAs is an ideal material system for eddy current sensing because of its high liquid electrical conductivity and large liquid:solid conductivity ratio. For semiconductor systems of similar (or higher) conductivity, eddy current data collected at around 500kHz -1MHz with either the absolute or the differential sensor schemes can be used to locate the position of solidification to better than ±0.5mm. Less ideal systems such as CdTe require the exploitation of fringe fields at the ends of the excitation coil together with a differential sensing scheme to achieve similar location precision. Once the interface’s location is obtained, lower frequency data can be used to deduce the interface’s curvature with acceptable precision for both material classes.

In order to validate the eddy current sensor concepts and analysis strategies presented thus far in a vertical Bridgman crystal growth application, a high temperature
eddy current sensor was installed in a commercial six-zone Bridgman furnace. Chapter 8 describes in detail the design, installation and testing of an “absolute sensor” for continuous monitoring of Cd$_{1-x}$Zn$_x$Te single crystal growth.
Chapter 8: Monitoring Bridgman crystal growth

8.1 Introduction

The yield and quality of material grown by the Bridgman technique is maximized by empirically optimizing the temperature gradient and the axial translation rate for each material system. Essentially, this involves repeated experimentation until a satisfactory material can be grown. Once obtained, temperature set points within the furnace, and furnace translation rate schedules are rigidly controlled from run to run. For high purity materials which have a low thermal conductivity (e.g. such as CdTe), it can be difficult to enforce the optimal thermal field during crystal growth and to ensure that solidification initiates and continues at the best location within the temperature gradient of the crystal grower. Reports of low substrate yields from crystals grown by this process may have arisen in part from a variability in the interface position (with respect to the furnace’s thermal gradient) and the development of non-ideal interfacial shapes during growth. The most critical parameters of the growth process to measure include the melt’s composition (i.e. its Cd:Te ratio) and temperature, the nucleation of solid on cooling, the liquid-solid interface velocity (i.e. the interface’s position within the ampoule as a function of time), the interface’s curvature (which affects the probability of secondary grain nucleation and successful competitive growth) and the temperature within the cooling solid.

Here the “absolute” sensor design has been incorporated into the ceramic liner of a six zone commercial vertical Bridgman furnace and several Cd$_{0.96}$Zn$_{0.04}$Te growth runs monitored. The resulting data is used to determine the melt stoichiometry, to detect the location of the liquid-solid interface, infer its curvature and to monitor the post solidification cooling of the solidified ingot. The study reveals that the Cd$_{0.96}$Zn$_{0.04}$Te vertical Bridgman growth process studied here probably occurs from a Cd depleted melt with a far from ideal liquid-solid interface shape.
8.2 The vertical Bridgman furnace.

A six zone vertical Bridgman furnace used for the commercial production of large (up to 3.5kg) CdZnTe ingots was used for an investigation of the eddy current sensing of vertical Bridgman crystal growth, Fig. 8.1. The furnace, Fig. 8.1(a), consisted of an assembly of three modular Marshall-type tube furnaces manufactured by Thermcraft Inc. The geometry and temperature capabilities of these modules are given in Table 8.1. The furnaces utilized resistance heating by internally exposed 0.204" diameter Kanthal wire elements helically wound at three turns per inch. A cylindrical alumina liner was placed inside the furnace to eliminate direct radiation transport from the exposed windings to the quartz ampoule used to contain the Cd$_{1-x}$Zn$_x$Te charge.

![Diagram](image)

Figure 8.1 a) Schematic diagram of six zone vertical Bridgman furnace. b) Furnace temperature profile and measured temperatures within an empty ampoule.

The top (#1) furnace was operated as a separately controlled, single zone furnace. The middle (#2) and lower furnace (#3) units were set up as three and two zone furnaces respectively. They were equipped with independent temperature controllers to establish the axial temperature profile needed for crystal growth.
Table 8.1: Characteristics of the Marshall Furnace Modules.

<table>
<thead>
<tr>
<th>Furnace No.</th>
<th>Model No.</th>
<th>Size (Length x Outer Diameter)</th>
<th>Rated Power (W)</th>
<th>Maximum Service Temperature (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1243</td>
<td>56cm x 36cm</td>
<td>4563</td>
<td>1316</td>
</tr>
<tr>
<td>2</td>
<td>1243-S</td>
<td>38cm x 36cm</td>
<td>2988</td>
<td>1316</td>
</tr>
<tr>
<td>3</td>
<td>1243-S</td>
<td>38cm x 36cm</td>
<td>2988</td>
<td>1316</td>
</tr>
</tbody>
</table>

Six thermocouples (TC 1-6) were positioned within the heating elements of each furnace zone and were used to control their temperatures (see Fig. 8.1(a) for their approximate locations). The temperatures of all six zones of the furnace assembly were independently controlled by a Model 823 MicRicon programmable process controller. The furnace was also equipped with two additional thermocouples (TC 7 and 8) located in the furnace wall. TC 7 was located between zones 3 and 4 while TC 8 was positioned between zones 5 and 6, Fig. 8.1(a). The temperatures at each thermocouple location are shown in Fig. 8.1(b). During a growth run, the furnace temperature at each thermocouple location was held constant while the furnace was translated vertically through the movement of a lead screw driven by a Compumotor Model SX-6 stepper motor. The position of the furnace was conveniently referenced by the location of a pointer (attached to the moving furnace) relative to a fixed measuring rule attached to the supporting framework, Fig. 8.1(a).

Prior to each growth run, the axial temperature profile along the furnace axis was measured with an array of nine R-type thermocouples spaced 5.1cm apart along the central axis of an empty quartz ampoule. Fig. 8.1(b) shows an example of this profile (together with the eight furnace thermocouple temperatures) for a pointer position of 14.9cm. Obviously, the axial temperature profile within the stationary ampoule will be dependent on the position of the furnace which varies with time during a growth run. To characterize this, the furnace was first moved to its start position (a pointer reading of
5.1 cm), a R-type thermocouple was then positioned at the bottom of an empty ampoule and a temperature measurement made once thermal equilibrium had been achieved. The thermocouple was then raised 25.4 mm to a new axial location and a new reading obtained. This process was repeated until a distance of 178 mm (which covers the ingot lengths typically grown in this furnace) had been profiled. After the measurements were completed with the furnace at its start position, the furnace was moved upwards by 25.4 mm to a new location and a new axial temperature profile measured as before. These temperature measurements were repeated until the full furnace translation used for the growth runs had been covered. Fig. 8.2 shows these temperature profiles together with the location of the sensor relative to the ampoule tip. This data is sufficient to characterize the temperature within the region of an ampoule interrogated by the sensor.

![Figure 8.2 Measured temperature profiles inside an empty ampoule for varying positions of the furnace.](image-url)
8.3 Sensor design and measurement methodology.

The basic sensor design analyzed in earlier studies consisted of a seven turn primary solenoid and a one turn secondary of slightly smaller radius located midway along the primary's length. To minimize perturbations to the temperature profile of the furnace, the two eddy current sensor coils were wound on the exterior and interior surfaces of a section of the alumina liner that is normally located in the annular space between the furnace walls and the ampoule containing the sample, Fig. 8.3. This enabled the installation of the sensor inside the furnace without significantly perturbing the thermal environment during the growth runs. The liner assembly consisted of three separate alumina tube pieces recessed for ease of insertion and removal of the sensor. Seven turns of 18 gauge platinum wire with a 6.35mm spacing were used for the primary (driver) coil. The "pickup" coil consisted of a single turn of 30 gauge platinum wire centered on the primary coil. In order to constrain the movement of the wires during heating/cooling, a high temperature cement (Saureisen Cement No.8) was applied to both sets of coils.

![Sensor Design Diagram](image)

Figure 8.3 A two coil eddy current sensor integrated into the alumina liner of the furnace.
Approximately 0.9m long sensor leads sheathed within high temperature Nextel sleeves connected the sensor coils to 1.2m long coaxial cables leading to the measurement instrumentation.

A schematic of the eddy current sensor measurement system is shown in Fig. 8.4. A continuous signal to the primary coil was provided by the variable frequency oscillator within a Hewlett Packard HP4194A Impedance Gain/Phase Analyzer. This signal was amplified with a Model 25A100 Amplifier Research Inc. amplifier to increase the field strength of the primary coil. Frequency dependent gain measurements (G) for the two coil system were made by recording the ratios of the voltage induced across the secondary coil and the voltage drop across a 1 ohm low inductance precision resistor in the primary circuit. The phase difference (\( \phi \)) between these two voltage signals was also monitored. Using the expressions in Fig. 8.4, the real and imaginary components of the sensor’s transfer impedance, Z, was calculated from the gain and phase. To emphasize the effects of the sample on the transfer impedance, the impedance was normalized by that of the empty (i.e. no sample) sensor. These “empty sensor” gain/phase measurements (\( G_e/\phi_e \)) were made at the growth temperature since they depend on the (temperature dependent) dimensions of the sensor.

### 8.4 Sensor calibration

When an eddy current sensor is installed within a crystal growth furnace, the eddy current sensor interacts with the sample and surrounding furnace. In addition, the long lead lengths between the sensor coils and the measurement instrumentation potentially introduce “anomalous” contributions to the measured impedance. Thus, a calibration of the sensor was performed using a 75mm diameter, 152mm long silicon cylindrical sample (Lattice Materials Corporation) contained in a quartz ampoule within the growth furnace, Fig. 8.5(a). Four point probe resistivity measurements performed on the silicon ingot indicated a conductivity of 2994 S/m. The experimental multifrequency normalized impedance curve for this silicon sample is shown in Fig. 8.5(b) for a frequency range of...
50kHz-3MHz. Standard eddy current analysis methods[150] were applied to this data and led to a deduced electrical conductivity of 2150 S/m for a test frequency near the knee of the normalized impedance curve (~267 kHz).

Figure 8.4 The eddy current test setup using a HP 4194A impedance analyzer to measure the multifrequency gain/phase response of the sensor. Data is normalized by the empty coil's response and the resulting complex transfer impedance plotted on an impedance plane diagram.

The standard analysis technique assumes infinitely long samples and solenoids[150]. Clearly, this would not be the case here (see Fig. 8.5(a)). To determine and correct for the effects of both the finite sample (i.e. edge effects) and the finite coil (i.e. fringe field effects) lengths, a finite element model was created for a 75mm diameter, 152mm long silicon sample located in a seven turn sensor of the geometry used here. The positions of the sensor coils (primary and secondary) relative to the location of the silicon sample are shown in Fig. 8.5(a). The furnace hardware was not modelled in the analysis. The transfer impedance of the sensor was calculated at 11 selected frequencies between 50kHz and 3MHz with a sample electrical conductivity (specified in the finite element
model) of 2994 S/m. The standard analysis method when applied to synthesized impedance data led to a recovered electrical conductivity of 2200 S/m very close to that measured with the eddy current sensor. Thus, the finite lengths of sensor/sample result in a ~26% underestimate of the true conductivity when a “standard” conductivity analysis is used[153]. A correction factor was therefore applied to the conductivities deduced during the monitored growth runs.

For this calibration, the conical shape of the bottom of the quartz ampoule resulted in one end of the cylindrical silicon sample being only 35mm away from the center of the sensor (or 15mm from the bottom of the excitation (primary) coil), Fig. 8.5(a). During a growth run, the molten charge obviously fills the cone shaped region, and so the edge effects for the Cd$_{1-x}$Zn$_x$Te sample might be slightly smaller than they were for the silicon sample.

Figure 8.5 Measured and calculated sensor responses for a silicon calibration sample.
The furnace was observed to have little effect upon the sensor’s response except in the 150kHz-250kHz range. The significance of this 150kHz-250kHz perturbation varied as the furnace was translated relative to the sensor. The smallest (an almost negligible) effect was observed when the 5cm insulation layer between furnace modules 2 and 3 was aligned with the center of the sensor. This corresponded to the situation where the furnace windings and metal containment were furthest from the pickup coil of the sensor. The measurements with the calibration silicon sample, Fig. 8.5(a), were conducted with the furnace position adjusted to align the insulation layer with the sensor.

8.5 Samples for growth runs.

Two crystal growth experiments (run #s 862 and 869) were monitored with the eddy current sensor. The samples were prepared from pieces of pre-compounded CdZnTe material previously synthesized using in situ distilled, zone refined, high purity grade (99.9999% or better), elemental Cd, Zn and Te. For the first run (#862), a charge weighing 3020g and having a 4.0% (atomic) Zn content was used. For the second experiment (#869), the charge weighed 2948g and contained 4.25% (atomic) Zn. The slight difference in Zn concentration has a negligible effect upon the electrical conductivity. About 3.8g of excess Cd was added to the charge to compensate for Cd evaporation into the ampoule’s large free volume. The charge material was contained in a 72.5mm inner diameter x 74mm outer diameter x 190mm long pyrolitic boron nitride (pBN) crucible (conically shaped at one end), placed within a 76mm inner diameter x 80mm outer diameter x 90cm long, semiconductor grade quartz ampoule. The ampoule was evacuated to 10⁻⁶ torr and sealed (with a hemispherical quartz cap to an effective inside evacuated length of 84cm). The ampoule was supported in the furnace by a set of concentric mullite tubes machined to different lengths to accommodate the conically shaped bottom end of the ampoule.
8.6 Growth experiments

For both of the growth experiments, the furnace was initially located at a relative start position of 5.1 cm, Fig. 8.1(a). A seventeen segment furnace heating, translation and cooling schedule was programmed in the Micron process controller. The first six segments caused the furnace temperature to be increased over a period of approximately 4 hours in order to melt the charge. At the completion of the sixth segment, the furnace temperature set points in zones 2 thorough 6 were 1137°C, 1133°C, 1133°C, 1020°C and 800°C respectively. During this four hour heating period, the cadmium over pressure region (zone #1) was simultaneously increased to a set point of 825°C. Once these temperatures were reached, a two hour soak (segment #7) was used to allow the melt to reach equilibrium before beginning furnace translation at 1.475 mm/hr (segment #8) whilst maintaining each zone at its fixed set point temperature. The programmed segments for solidification and subsequent cooling (segments 8-17) are summarized in Table 8.2.

<table>
<thead>
<tr>
<th>Seg #</th>
<th>Activity</th>
<th>Duration(hrs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>Move furnace up at 1.475 mm/hr</td>
<td>13.3</td>
</tr>
<tr>
<td>9</td>
<td>Stationary furnace</td>
<td>6.0</td>
</tr>
<tr>
<td>10</td>
<td>Move furnace up at 1.475 mm/hr</td>
<td>13.3</td>
</tr>
<tr>
<td>11</td>
<td>Stationary furnace</td>
<td>6.0</td>
</tr>
<tr>
<td>12+13</td>
<td>Move furnace up at 1.475 mm/hr</td>
<td>76.7</td>
</tr>
<tr>
<td>14</td>
<td>Stationary furnace, cool down to 1090°C</td>
<td>4.0</td>
</tr>
<tr>
<td>15</td>
<td>Move furnace down at 5.9 mm/hr and cool down to 1025°C</td>
<td>21.6</td>
</tr>
<tr>
<td>16</td>
<td>Stationary furnace, cool down to 825°C</td>
<td>20.0</td>
</tr>
<tr>
<td>17</td>
<td>Stationary furnace, cool down to 625°C</td>
<td>10.0</td>
</tr>
</tbody>
</table>
During the crystal growth experiments, eddy current sensor gain/phase data was collected (from the beginning of segment #8) at 101 logarithmically spaced test frequencies between 50kHz and 6MHz. A total of 256 samples were averaged with an integration time of 5ms for each sweep frequency resulting in approximately a five minute data collection period. Due to the slow furnace translation rate (1.475mm/hr), the data was collected and downloaded to a personal computer once every 10 minutes throughout the monitored growth and cooling periods (140-180 hours).

After the growth period (i.e. at the end of segment #13), segment #14 allowed the temperature of zones 3 and 4 to initially cool to 1090°C over a 4 hour period. The furnace was then translated downwards (during segment #15 over a 21 hour period) to an annealing position with the temperature reduced further to 1025°C. With the furnace stationary at the end of segment #15, a controlled cool down to 825°C was scheduled over 20 hours (segment #16), followed by a 10 hour cool down period to 625°C (segment #17) before the furnace power was turned off for natural cool down to room temperature.

Furnace equipment malfunctions during the growth runs resulted in several unintended changes to the originally programmed segments. In the first experiment, a thyristor failure in the last stages of segment #15, curtailed the segment to 18.2 hours (from 21.6 hours) and was followed by an intentional jump to segment #17. Since this disrupted the desired annealing schedule, impedance data collection was terminated at the end of segment #15. In the second experiment, a change had to be made in segment #12 following the discovery that the furnace had failed to translate for an indefinite time (during an overnight period). The furnace translation motor was reset and additional time programmed in to segment #12 to compensate for the unintentional stationary period (of the furnace) and thus allow the furnace to eventually move over the full growth distance.

Post growth analysis on the two ingots from runs #862 and #869 were conducted to evaluate the infrared transmission properties (FTIR), the existence of Te precipitates, and the defect density (EPD). These findings are summarized in Table 8.3[166].
Table 8.3: Post growth characterization data from ingot #862 and #869[166].

<table>
<thead>
<tr>
<th>Property</th>
<th>#862</th>
<th>#869</th>
</tr>
</thead>
<tbody>
<tr>
<td>FTIR (2 - 16 μm)</td>
<td>&gt; 60 %</td>
<td>&gt; 60 %</td>
</tr>
<tr>
<td>Te precipitates</td>
<td>&lt; 2 μm</td>
<td>&lt; 2 μm</td>
</tr>
<tr>
<td>EPD (cm⁻²)</td>
<td>2E4</td>
<td>4E4</td>
</tr>
<tr>
<td>Zn % (measured)</td>
<td>4.17</td>
<td>3.3</td>
</tr>
</tbody>
</table>

8.7 Results and discussion

The complex normalized impedance at 101 logarithmically spaced frequencies between 50kHz and 6MHz was calculated from the measured gain and phase data. Fig. 8.6(a) shows the imaginary component of impedance (normalized by that of the empty sensor at the growth temperature) for four frequencies as a function of time for the first experiment (run #862). The time axis began when the furnace commenced its upward translation from its 5.1cm start position (i.e. the start of segment #8). The shaded areas in Fig. 8.6(a) indicate segments during which the furnace was held stationary. The first two pause segments (#9 and #11) were included with the intention of controlling undercooling and to help initiate nucleation at the early stages of the run. For run #862, the growth period was 115 hours and was followed by an annealing period of approximately 26 hours.

Using the furnace temperatures and the profiling data of Fig. 8.2, the temporal variation of the temperature near the sensor was estimated. Fig. 8.6(b) shows the time variations of temperature near the furnace liner (where the sensor was located) and at the center of an empty ampoule. This latter temperature is of course only an estimate of the exact temperatures within the sample and depends on the thermal transport properties of the solid and liquid. For low thermal conductivity materials like Cd₁ₓZnₓTe, significant radial temperature gradients are expected to be present in the large diameter boules used here.
Figure 8.6 a) Measured impedance response at four frequencies, b) Furnace temperature variation, and c) "Apparent" electrical conductivity variation during crystal growth and cooling for run #862.
From the time varying complex impedance data, the variation in the "nominal" conductivity was calculated using a standard (infinite sample/infinite sensor) analysis [153]. The "nominal" electrical conductivity variation deduced from the measured impedance data was then corrected for the finite length sample/coil effects (from the finite element modelling study) and plotted in Fig. 8.6(c) as the "apparent" conductivity. We use "apparent" because the conductivity observed during the time period when the sensor interacted with both solid and liquid is ill-defined.

Figs. 8.7(a)-(c) show the results obtained from the second experiment. The starting position and furnace translation rate were the same as for the first run. The impedance and conductivity changes and their trends observed from this experiment were very similar to the first experiment. One significant difference arose from the unintended pause in the furnace translation; this extended the growth period to 130 hours. It was also possible to monitor the annealing segments more fully; cooling data for this run was obtained over the longer period of 46 hours.

8.7.1. Melt characterization

The results of Figs. 8.6 and 8.7 show that the change of state from a liquid to a solid state causes an attendant decrease in electrical conductivity and results in an increase in the imaginary impedance component towards unity (the empty sensor's value) for all of the test frequencies. During the first 60 hours of furnace translation, the high frequency imaginary impedance component was around 0.6 for both samples, consistent with the sensor's interrogation of a relatively good electrical conductor. The changes in impedance throughout this first 60 hours were relatively small, suggesting that the liquid's conductivity changed only moderately and the liquid-solid interface was too distant from the sensor to be detected. Finite element modelling results also indicated that the sensor interacted only with the liquid in this configuration. Thus, we conclude that the sensor
Figure 8.7 a) Measured impedance response at four frequencies, b) Furnace temperature variation, and c) "Apparent" electrical conductivity variation during crystal growth and cooling for run #869.
"sampled" only the liquid state and the data can therefore be used to infer the liquid's electrical conductivity.

The charge's liquid conductivity at the starting furnace location was found to be around ~18,000 S/m for both of the growth runs. This value of conductivity is significantly higher than that expected (7200-8000 S/m) for a stoichiometric Cd$_{0.96}$Zn$_{0.04}$Te melt even if it has been heated 30-40°C above its melting temperature[151]. Investigations of the effects of melt stoichiometry upon the electrical conductivity of binary Cd-Te and ternary Cd$_{1-x}$Zn$_x$Te alloys have indicated significant increases in conductivity for both Cd rich and Cd depleted melts[108,151]. The observation of an initially high conductivity here strongly suggests the existence of a non-stoichiometric melt but the data alone cannot distinguish between either Cd rich or Cd depleted. However, observations of tellurium precipitates after growth[166] in material grown in this furnace using similar growth run histories strongly suggests that these were Cd depleted melts.

The melt appeared to remain non-stoichiometric for much of the growth. For example, after about 60 hours, the electrical conductivity had only decreased to ~12,000S/m. It is therefore concluded that the liquid was significantly depleted in Cd from the start of furnace translation until at least the time when the solidification front reached the sensor.

8.7.2. Interface characterization

As the liquid-solid interface approached the sensor (after ~60 hours for run #862 and ~75 hours for run #869), a gradually increasing fraction of the low conductivity solid appeared in the sensor's "field of view" and the imaginary impedance was observed to rise for all of the frequencies measured, Figs. 8.6(a) and 8.7(a). This was consistent with the behavior predicted using finite element modelling in which the greatest change of impedance occurs when the interface passes roughly through the center of the sensor. The impedance data exhibited inflection points around 84 hours after the start of furnace translation for run #862 and therefore roughly indicated the time of passage of the solid-
liquid interface through the central region of the sensor. A time delay in the occurrence of the inflection point was observed in the second experiment (run #869). This was caused in part by the unintended time interval during which the furnace had not translated in segment #12. As a result, the interface appeared to have passed through the sensor after about 92 hours (Fig. 8.7(a)). As the interface moved upwards away from the sensor, a decreasing fraction of the liquid and an increasing fraction of the solid were sampled, and the impedance approached an asymptote determined by the conductivity of the solid material.

The time at which the inflection point was observed has been predicted to depend on the interface location, its curvature and the test frequency. The modelling study indicated that the time at which the inflection point would be observed increases as the curvature changes from convex interface to flat, to concave, Fig. 8.8(a). Fig. 8.8(b) shows the effect of test frequency upon the inflection point as a flat interface passed through the sensor. The modelling study revealed that the inflection points at higher frequencies became more independent of the interface shape and converged to a position that corresponded to near alignment of the outer edge of the interface with the sensor's midpoint, Fig. 8.8(c). This phenomenon potentially enables the (interface shape independent) determination of the location and thus the growth rate of the interface. Once the time at which the interface was located at the center of the sensor was determined (h=0), the relative large shifting of the inflection points at lower frequencies (shown in Fig. 8.8(c) for CdTe) could then be used to estimate the shape of the interface (convex, concave or flat).

Figs. 8.9(a) and (b) show the frequency dependence of the inflection point time for runs #862 and #869 respectively. From the high frequency data, the time at which the interface was located at the center of the sensor was found to be ~84 and ~92 hours for run #862 and #869 respectively. If it is assumed that there is no delay in nucleation upon the start of translation, that the interface was flat, and that the movement of the interface (i.e. the growth rate) equalled the furnace translation rate, the time taken for the interface to
reach the position of the sensor would be 66 hours for the first experiment (run #862) and 81 hours for the second (run #869). These are significantly less than those measured (84 and 92 hours respectively). The difference in the estimated and actual times could be due to 1) a growth rate not equal to the furnace translation rate, 2) a non-planar interface shape and 3) a delay in the onset of nucleation as a result of liquid supercooling. If the third possibility is discounted and we assume a shape with the convexity deduced below, the eddy current observed times for the passage of the interface through the sensor imply actual growth rates of 1.105 mm/hr for the first experiment and 1.244 mm/hr for the second. In previous experimental work with CdTe, growth rates of less than 35% of the translation rate have been reported[167]. The rates of 1.105 mm/hr and 1.244 mm/hr are less than the furnace translation rate (1.475 mm/hr) and so solidification occurred lower in the furnace than anticipated.

![Diagram](image)

**Figure 8.8 a)** Calculated impedance variation with interface position for three interfaces.
Figure 8.8 b) Calculated impedance variation with interface position for a flat interface at four frequencies.

Having determined the moment when the interface was located at the sensor's center, it is possible to use the result from the previous modelling study (Fig. 8.8(c)), to determine the interfacial curvature. Fig. 8.10 plots the frequency dependence of the inflection point (relative to the interface location) against frequency for the two experiments, compared with the calculations done for the three interfaces shown in Fig. 8.8(c). In Fig. 8.10, the flat interface curve of Fig. 8.8 has been replaced by a calculated curve accounting for the reduced diameter of 72.5mm (of the pBN crucible) and actual liquid/solid conductivities deduced from the growth runs. It is apparent from this data that the interface curvature lay between the convex and flat cases modelled (Fig. 8.8(c)). The low frequency data for both growth runs was similar indicating a convexity of about 0.167. Small differences in the high frequency (small skin depth) eddy current behavior of the samples are indicative of a greater flattening of the interface near the ampoule wall for run
The recovery of a more exact shape of the interface requires a detailed multifrequency inverse analysis of the impedance data.

Figure 8.8 c) Calculated position of the inflection point vs. frequency for three interfaces.

Previous modelling work has shown that for material systems with liquid:solid thermal conductivity ratios (k_l/k_s) greater than one, the interface shape is concave in the presence of a uniform temperature gradient[168]. The interface could be made convex by reducing the temperature gradient above the interface and/or increasing the temperature gradient below the interface[167,169]. Evidence of a non-stoichiometric melt suggests solidification to be occurring at a lower temperature than 1099°C-1100°C (the liquidus of stoichiometric Cd_{0.095}Zn_{0.045}Te) and therefore consistent with the observation of the solidification front located at a lower than anticipated position in the moving furnace. In this region of the furnace the temperature gradient is significantly steeper (Fig. 8.1(b)), and may account for the less than ideal shape of the interfaces.
Figure 8.9  a) Frequency dependence of the time appearance of the inflection point for run #862. b) Frequency dependence of the time appearance of the inflection point for run #869.

Figure 8.10 Comparisons of experimental and calculated variations of the inflection point position with frequency.
8.7.3. Solid state annealing/cooling

Proper post growth annealing/cooling assists in the homogenization of the Zn distribution and controls the defect density and the concentration of Te precipitates. The defect density and number of precipitates in a grown boule are affected by the cooling rate and the overall cooling period. During cooldown, the impedance data collected from the eddy current sensor can be used to evaluate the conductivity of the cooling crystal and assess its potential for characterizing some aspects of post-growth phenomena.

For the first experiment, a solid conductivity of 923 S/m was observed at the end of growth (segment #13), which further decreased to 791 S/m during a four hour stationary furnace segment (#14) when the furnace zone temperatures in zones 2 through 4 were reduced by approximately 48°C. In the next segment (#15), the furnace was moved down to an annealing position, while the temperatures in zones 2 through 5 were decreased further by approximately 53°C. During these two segments, the temperatures in zones 1, 5
and 6 were maintained at 825°C, 1020°C and 800°C respectively. Data analyzed from the second experiment (run #869) revealed similar results. In this case, a post growth conductivity of 839 S/m was observed which decreased to 712 S/m during the initial four hour cooling period (segment #14).

An anomalous increase in sample conductivity has been detected during segment #15 when the furnace was moved to its annealing position. A small increase in conductivity with decrease in temperature was detected in prior experiments with Cd$_{0.96}$Zn$_{0.04}$Te samples used to characterize the electrical conductivity variation of Cd$_{0.96}$Zn$_{0.04}$Te with temperature[150,151]. In those experiments, a slight anomalous increase in conductivity occurred after solidification was initially detected (i.e. within 40°C of solidification) and was then followed by a gradual decrease in conductivity with temperature during the remaining cool down. This anomalous behavior was not observed in two other experiments conducted with CdTe and Cd$_{0.92}$Zn$_{0.08}$Te[150,151]. For these compositions, the solid conductivity was 20-30% lower and decreased monotonically with decreasing temperature, Fig. 8.12. Modelling studies of the Zn segregation has revealed concentrations varying from ~5.8% (near the first to freeze region) to 2.8% (near the top of the boule)[30]. One explanation for the rise in conductivity during post growth cooldown is the homogenization of Zn by diffusion which results in a more uniform concentration closer to 4% with an increased conductivity. In Run #869, with the furnace stationary, segment #16 allowed a controlled furnace cool down to 825°C during which time a gradual decrease in conductivity was observed consistent with the decrease in temperature.
Figure 8.12 Electrical conductivity variation of Cd$_{1-x}$Zn$_x$Te with temperature for $x=0.0$, 0.045 and 0.08[150,151].

8.8 Summary

Detailed experiments have been conducted with an eddy current sensor in a vertical Bridgman furnace to better understand the CdZnTe growth process and to obtain information about the solid-liquid interface. From the measured impedance data, the sensor has demonstrated the capability of providing significant new information about the melt composition and has revealed that growth occurred from a non-stoichiometric (probably) Cd depleted melt. An analysis of the high frequency data has enabled the interface’s location to be deduced and has revealed that the growth rates were lower than the furnace translation rate so that solidification occurred in a colder section of the furnace where the temperature was considerably steeper than anticipated. Lower frequency data
revealed that growth occurred with a less than ideal convex interface shape. Post growth monitoring of the cooling process has enabled the electrical conductivity data of the solid to be monitored. An "anomalous" increase in conductivity was observed as the temperature was decreased from the solidification temperature, perhaps as a result of Zn homogenization.
Chapter 9: Discussion

The inability to obtain a high yield of good quality $\text{Cd}_{1-y}\text{Zn}_y\text{Te}$ single crystals on a consistent basis has led to the development of non-invasive eddy current sensors for vertical Bridgman crystal growth. A high temperature sensor installed in a commercial Bridgman furnace has been able to characterize the melt (in terms of its electrical conductivity), determine the interface position and solidification velocity, and characterize the interface shape.

The successful application of this sensor technique depends on a relatively large electrical conductivity change accompanying melting or solidification which is a common feature of most semiconductors. For compound semiconductors, the electrical conductivity also exhibits a compositional dependence. It is necessary to have well established electrical conductivity-composition-temperature relationships to enable the successful implementation of eddy current sensors for monitoring crystal growth. In $\text{Cd}_{1-y}\text{Zn}_y\text{Te}$ alloy crystal growth, Cd evaporation affects the composition of the melt. By monitoring the electrical conductivity with the eddy current sensor, it has been possible to characterize the initial melt state and identify whether growth occurred from a stoichiometric or non-stoichiometric melt.

Crystal growth rates were found to be lower than the furnace translation rates. The onset of nucleation was not detectable with the remote placement of the existing sensor but has been observed in recent work of Choi[151]. Since a convex liquid-solid interface shape was revealed, a shorter time for the passage of the interface through the sensor was expected. However the observed average slow growth rate is an indication that there was a delay in the onset of nucleation. It is clearly necessary to identify this possibility and it is proposed that an additional sensor should be installed near the ampoule tip to characterize supercooling/nucleation phenomena. Another intriguing prospect arising from the placement of an eddy current sensor at the ampoule tip for seeded Bridgman growth, is the possibility of monitoring seed melt back for process design optimization and real-time control. Differential sensor techniques appear to have merit in this application.
The eddy current sensor can also be used during post growth cooling/annealing to closely monitor the electrical conductivity history. For this, detailed studies of the conductivity-temperature relationship and phase transformation kinetics are required[150,151].

The feasibility of an eddy current sensor approach to crystal growth was first investigated using axisymmetric electromagnetic finite element models for two reasons. One reason is the fact that a crystal growth run typically takes 7-10 days for completion, which is a time consuming and expensive way of evaluating suitable sensor designs. The second reason for using electromagnetic finite element models and analysis tools is the design flexibility it provides in selecting an appropriate sensor design for a particular materials system. The electromagnetic interactions between a chosen sensor design and a sample can be studied in detail by performing multifrequency simulations. A comparison of candidate designs can be made on the basis of the materials system (selection of test frequencies) and the physical geometry (sensor and sample sizes).

The imaginary impedance component vs. frequency plots were found to be useful for comparisons due to the monotonic behavior (increase) of the imaginary impedance during a liquid to solid transformation. At high frequencies the interface position had a greater influence on the impedance response than the interface shape. It was observed that the sensitivity to a moving interface was highest for the materials system with the largest liquid:solid electrical conductivity ratio. In addition, the sensitivity showed a frequency dependence which was based on the liquid and solid conductivities and the different electromagnetic depths of penetration (skin effect) into each material. Thus, absolute values of liquid and solid conductivities and their ratios were found to be important parameters in determining the operating frequency ranges best suited for each material system.

A separate experimental study on a model system provided insight to the contributions to the sensor response from parasitic circuit effects. For lower conductivity systems, inclusion of a circuit analysis to complement the electromagnetic analysis
appears necessary if interpretations of the interface position and shape are made from the imaginary part of impedance vs. frequency plots. This requirement can be deferred by using a separate analysis protocol to recover interface position and shape from the behavior of the sensor response curves as the interface passed through the sensor. For the absolute sensor, the continuous tracking of an inflection point in the response curve at high frequencies, was found to be a method of determining the growth rate. The low frequency analysis of the inflection point data indicated possibilities of characterizing the shape. For the differential sensor design, analysis of the appearance of a peak in the sensor response resulted in a similar scheme of identifying the position (high frequency) and shape (lower frequency) of the interface.

The two sensors analyzed in this research are not optimum designs. For the multiple turn primary coil, the coil diameter, the number of turns, the spacing between individual turns, the overall coil length, the energizing current and the test frequency are design parameters that affect the electromagnetic field. The reduction of the number of turns/coil length (in the limit a single turn) in the primary will result in fringing fields having a bigger effect on the sensor response, more so for differential designs. These have greater sensitivity, but produce weaker signal levels. For the differential sensor, the placement and the separation distance between the secondary coil turns provides an additional design variable. Geometrical constraints affecting design selections include the available annular space between the furnace wall and the ampoule (affecting the choice of primary and secondary coil diameters), the charge weight and resulting length of the ingot (affecting the sensor length, number of turns, coil spacing), the shape of the ampoule (for example, cylindrical vs. conical shaped sensor designs at the ampoule tip). Incorporation of sensors to existing components of the furnace assembly (e.g. liner) may be a necessity to minimize effects of growth conditions (e.g. temperature profile). A more comprehensive evaluation of parametric sensor designs needs to be done.

Thermal models of the Bridgman growth process reveal the temperature fields at different stages of growth. If a complete electrical conductivity-temperature relation is
available, the electrical conductivity field distribution can be used in the material property specification of each element. The existing models utilize conductivity values at the melting point (single values for the liquid and solid). A more true representation of the axial and radial temperature gradients can be used in the models by specifying space \((r,z)\) dependent conductivity values. This approach could provide a link between thermal models characterizing the interface and eddy current sensor models.

The eddy current sensing approach has shown considerable promise of providing important observations of the vertical Bridgman growth process. When properly implemented and carefully interpreted, they provide useful information to the crystal grower in striving to reach higher yields of better quality semiconductors. Prospects are encouraging for the implementation of an eddy current sensor based manufacturing approach for semiconductor crystal growth. One such arrangement is shown in Figure 9.1 to continuously track the liquid-solid interface using feedback control.

![Image](image_url)

**Figure 9.1** Eddy current sensor based solid-liquid interface tracking system
In terms of economic impact of using an eddy current sensing arrangement, the impedance analyzer should be replaced with a less expensive oscillator and a magnitude and phase measuring system (lock-in amplifier) to reduce costs. This could be a key element in the acceptance and application of eddy current sensors by the crystal growth community.
Chapter 10: Conclusions

Substrates for infrared sensitive focal plane arrays suffer from undesirable variability in properties, high cost and inadequate size. Concerted attempts have been made to improve the vertical Bridgman growth of bulk semiconductor crystals (from which, these substrates are obtained). This includes the development of mathematical models of the growth process and sensor techniques for characterizing the liquid-solid interface. The mathematical models developed have ranged from basic 1-D models to full 3-D models. Comparisons or verifications of these analytical models have been made difficult due to the varying assumptions being used in their formulations (e.g. the inclusion/exclusion of convection, latent heat, segregation).

Sensor based methods rely on the ability to measure changes in physical properties occurring during a liquid to solid transformation. For semiconductors, one such property is the electrical conductivity, which increases exponentially with increasing temperature and undergoes an abrupt change upon melting. For a growing crystal, the distribution of electrical conductivity is reflective of the local temperature and whether the region is liquid or solid. The electrical conductivity can be indirectly measured in terms of the electrical impedance of an eddy current sensor.

In the development of eddy current sensors, electromagnetic finite element analysis tools were used. The electromagnetic interactions between two chosen designs ("absolute" and "differential" sensors) and a sample were studied in detail by performing multifrequency simulations of different liquid-solid interface positions and shapes for a range of materials systems. It was concluded that by making multifrequency measurements, it was possible to use analysis protocols to determine the interface position and shape. The feasibility of the sensor concept was investigated in a model Silicon system and significant agreement was observed between the finite element model calculations and experimental results.
A high temperature sensor was designed, fabricated and installed in a commercial 6-zone vertical Bridgman furnace to monitor Cd$_{0.96}$Zn$_{0.04}$Te crystal growth runs. Analysis of the eddy current sensor data indicated a convex interface passing through the stationary sensor at slower growth rate than the furnace translation rate. It was also observed that the sensor could be used to monitor other aspects of the growth process such as characterizing the initial melt composition prior to growth, and conductivity measurements during post growth annealing. Additional sensors placed at the bottom of the ampoule should provide more information about the growth process such as supercooling, nucleation and seed melt back phenomena.

In conclusion, eddy current sensors have shown considerable promise of providing useful information to crystal growers to improve the vertical Bridgman crystal growth process. Prospects are encouraging for the implementation of an eddy current sensor based manufacturing approach for semiconductor crystal growth.
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