Effect of Magnetic Field in Bridgman Growth of Semiconductor Alloys

Th. Duffar, C. Stelian, A. Mitric

Abstract

The influence of an axial magnetic field on thermosolutal convection and chemical segregation during vertical Bridgman solidification of $Ga_{1-x}In_xSb$ alloys ($x = 0.001$ and $0.2$) has been investigated numerically by using the finite element software FIDAP® to compute the temperature, flow and solute fields in the samples. It was found that the $In$ rejected at the solid-liquid interface causes a stabilizing density gradient and rapidly damps the thermally driven convection. The damping of the melt convection causes an increase of the radial segregation at the beginning of the solidification. By applying an axial magnetic field of $B = 0.5 T$, the melt convection is further strongly damped and the radial segregation increases. On the contrary, it is expected that an increase of melt convection will decrease the radial chemical segregation in the crystals consequently. An experimental program of $GaInSb$ growth under rotating and sliding magnetic field has been defined.

Introduction

Concentrated semiconductor alloys, as for example $Ga_{1-x}In_xSb$, $Ga_{1-x}In_xAs$, $Ge_{1-x}Si_x$ and $Hg_{1-x}Cd_xTe$ are of considerable potential interest as their electronic properties can be adjusted by controlling the alloy composition $x$. However, the growth of such crystals is a formidable task because the high concentrations lead to unacceptable chemical segregation, associated misfit stresses and solid-liquid interface digging and destabilization. Taking $GaInSb$ as model material for this class of problems, numerical simulations of heat, momentum and mass transfer during transient solidification are performed in order to determine appropriate experimental growth conditions. They leaded to a good qualitative and quantitative understanding of the physics involved.

1. Bridgman configuration

A schematic diagram of the vertical Bridgman apparatus is shown in Fig.1a. The principle of Vertical Bridgman technique is to solidify a melted charge in a heat gradient. The crucible is pulled with the velocity $V$ along the $z$-axis of the furnace. The heater is a conical resistor that dissipates a longitudinally variable power. Several thermocouples placed along the crucible give the temperature field.

The curvature of the liquid/solid front and the reject of solute at the interface are the origin of axial and radial chemical segregations in the crystal. Extensive order of magnitude analysis and numerical analysis have shown that the radial segregation is sensitive to the $Pe$ and $GrSc$ numbers (see eq. 1) [1-3].
Fig. 1. a) Vertical Bridgman configuration; b) 2 photographic images of the solid-liquid interface shapes marked by electrical pulses, the sample diameter is 12mm; c) drawing of the marked solid/liquid interfaces.

Fig. 2 shows this variation. Some technical solutions can be used in order to solve, at least partially, these problems in the Bridgman configuration. It has already been shown that the axial chemical composition can remain essentially constant on a large part of the crystal provided that the liquid close to the solid-liquid interface is not mixed with the feeding liquid: for example by the use of baffles, or with a carefully controlled thermal field in the sample. A resulting improvement is that a constant concentration does not generate misfit stresses in the crystal. On the other hand a careful mixing of the liquid close to the interface is useful to stabilize the growth and also to prevent radial chemical segregation. Then the challenge is to provide a good mixing of the liquid close to the interface, but to prevent mixing of this layer with the remaining liquid.

We plan to investigate how the use of electro-magnetic fields during the solidification process can affect the results in term of chemical homogeneity. The Bridgman machine will be modified by adding a magnetic field. It is planned to grow GaInSb samples under various thermal and magnetic configurations, in order to measure, as a function of time, the thermal field, the solid-liquid interface velocity and shape and to analyze the radial and axial chemical segregation in the resulting ingots. The form of the interface and its rate of solidification are
obtained thanks to the marking of interfaces carried out at regular intervals (see Fig.1b and Fig.1c).

Experiments on \(Ga_{1-x}In_xSb\) \((x = 0.04, 0.1\) and 0.2) directional solidification involving the measurement of temperature distribution around the crucible, the interface curvature during the growth process and the solute distribution in the solidified samples have been performed by Duhanian [4]. The experimental results show a significant increase of radial segregations and interface deflection for the high-doped alloys (see Fig.1b and Fig.1c). In order to give a quantitative and qualitative explanation of these observations, a transient numerical simulation of the heat, mass and species transport during the concentrated alloys solidification is necessary.

2. Governing equations

The calculations presented here are limited to the crucible-sample system, which is assumed to exhibit symmetry around the \(z\)-axis. The convecto-diffusive movements within the fluid, in presence of a magnetic field, are described by the Navier-Stokes equation, the conservation of mass, energy and solute equations in the Boussinesq approximation including the electromagnetic interaction through the Lorentz-Laplace force.

The dimensionless numbers governing the problem are Grashof, Prandtl, Peclet, Schmidt and Hartmann numbers defined by:

\[
Gr = \frac{\rho^2 \beta_T \Delta T g R^3}{\mu^2}, \quad Pr = \frac{\mu c_p}{k}, \quad Pe = \frac{RV}{D}, \quad Sc = \frac{\mu}{\rho D}, \quad Ha = \frac{\sigma}{\mu} BR
\]

where \(\rho\), \(k\), \(c_p\), \(\beta_T\), \(\mu\), \(D\) and \(\sigma\) are the density, thermal conductivity, specific heat, thermal expansion coefficient, viscosity, diffusion coefficient and electric conductivity, respectively.

The dimensionless variables are defined by scaling length with the crucible radius \(R\), the velocity \(u\) by the characteristic speed \(U = \sqrt{\beta_T \Delta T g R}\), time with \(R/U\) and pressure with \(\mu U / R\). The dimensionless temperature is defined by \(\tilde{T} = (T - T_C) / \Delta T\), where \(\Delta T = T_H - T_C\) is the difference between the temperatures of the hot and cold zones of the furnace. The nondimensional solutal expansion is given by \(\tilde{\beta}_s = \beta_s / (\beta_T \cdot \Delta T)\).

The thermal boundary conditions are imposed on the external surface of the crucible using the temperature profile measured by thermocouples [4]. At the solid-liquid interface the energy balance includes the latent heat of fusion.

The no-slip condition on the velocity field is imposed at the solid boundaries of the liquid sample. For the applied magnetic field, the condition \(j = 0\), related to electric current density, must be satisfied at the crucible surfaces.

\[
(\frac{\rho}{\rho_s} \frac{1}{P_{elv}} \nabla \bar{C} \bigg|_{\text{liq}} - \frac{1}{P_{es}} \frac{D}{Dx} \nabla \bar{C} \bigg|_{\text{s}})n = (1 - K) \bar{V} \bar{C}_l
\]
3. Numerical simulation

The calculations presented here were performed with help of finite element code FIDAP® for \( Ga_{1-x} \text{In}_x \text{Sb} \) samples with radius \( R = 0.6 \text{cm} \) and length \( L = 6 \text{cm} \). This simulation concerns crystals grown in a three zone Bridgman configuration. The axial temperature gradient produced into the adiabatic zone of the furnace is \( G_T = 60 \text{K/cm} \) and the pulling rate of the crucible is \( V = 1 \mu \text{m/s} \).

The temperature, flow and solute fields computed for a diluted \( x = 0.001 \) alloy are shown in the Fig.3. The melt convection is characterized by two flow cells (see Fig.3b). The lower one is positioned near the solid-liquid interface and is driven by the radial temperature gradient caused by the interface curvature. The maximum value of the velocity, obtained on the symmetry axis is \( u_{\text{max}} = 4.5 \cdot 10^{-4} \text{m/s} \). This flow influences the solute distribution in the melt. As is shown in the Fig.3c, the isoconcentration lines are distorted in the vicinity of the interface. This characterizes a well-mixed regime in the melt.

Fig.3. Numerical results for \( Ga_{0.999} \text{In}_{0.001} \text{Sb} \) at time \( t = 1000 \text{s} \): a) thermal field; b) flow streamlines; c) solute field.

Fig.4. Velocity field in the melt for \( Ga_{0.8} \text{In}_{0.2} \text{Sb} \): a) \( t = 0 \text{s} \); b) \( t = 100 \text{s} \); c) \( t = 300 \text{s} \)
The modelling of the concentrated $Ga_{0.8}In_{0.2}Sb$ solidification shows a strong effect of the solute field on the melt convection. The $In$ rejected at the solid-liquid interface causes a stabilization of density gradient produced by the radial variation of the temperature in this region. This leads to a rapid damping of the convection, as is shown in the Fig.4. The center of the lower cell is moved to the crucible and the maximum velocity of the flow is reduced from $4.5 \cdot 10^{-3} m/s$ to $4.5 \cdot 10^{-5} m/s$ at time $t = 300s$. The isoconcentration lines become almost flat, which suggests that a diffusive regime is established in the melt. The radial segregation is caused now only by the interface curvature. In addition, as a result of the dependence of melting temperature on the interface composition, the melting point at the center of the sample decreases more than at the sides. This leads to an increase of the interface curvature during the solidification process (see Fig.5).

Fig.5. Solid-liquid interface shape for $Ga_{0.8}In_{0.2}Sb$ at time: A) 10s; B) 1000s; C) 2000s; D) 3000s

Fig.6. Radial segregation as function of solidified length

$Ga_{0.8}In_{0.2}Sb$ without magnetic field
$Ga_{0.8}In_{0.2}Sb$ without magnetic field
$Ga_{0.8}In_{0.2}Sb$ with magnetic field $B=0.5T$
The effect of an axial magnetic field on thermo-solutal convection is analyzed for the concentrated $Ga_{0.8}In_{0.2}Sb$ alloy. Application of a high magnetic field ($B = 0.5T$) generates Lorentz forces, which tend to reduce the convection intensity. From the numerical simulation, it is found that the flow cells are more damped and the center of the flow migrates toward the crucible wall. The influence of the applied magnetic field on the solute distribution is analyzed by computing the radial segregation $\delta C = (C_{\text{max}} - C_{\text{min}}) / C_{\text{av}}$ at the solid-liquid interface. The radial segregation versus the solidified length is plotted in Fig. 6. For the diluted $x = 0.001$ alloy growth without magnetic field, the radial segregation is low and almost constant. This can be explained by the high intensity of the thermo-convective flow, which mixes the solute rejected at the interface. In the case of concentrated alloy growth without magnetic field, the damping of the lower flow cell as a result of the solutal effect leads to a significant increase of the radial segregation. This is in agreement with Fig. 2: in the convective regime ($GrSc > 10^4$), if $GrSc$ is decreased, then $\Delta C$ increases.

In the case of concentrated alloy growth without magnetic field, the damping of the lower flow cell as a result of the solutal effect leads to a significant increase of the radial segregation. This is in agreement with Fig. 2: in the convective regime ($GrSc > 10^4$), if $GrSc$ is decreased, then $\Delta C$ increase. By applying an axial magnetic field $B = 0.5T$ in the case of the $Ga_{0.8}In_{0.2}Sb$ solidification, the convection is more damped and the radial segregation increases.

Conclusions

A numerical modelling of the concentrated $Ga_{0.8}In_{0.2}Sb$ alloy solidification has been carried out. The influence of high axial magnetic field on the thermo-solutal convection and solute distribution in the sample was numerically investigated and it is shown that, for an axial magnetic field of 0.5T, the melt convection is damped and the radial segregation increases. In order to decrease the radial segregation it is then better to increase the convective flow in the melt by using a rotating or sliding magnetic field. As a consequence, $Ga_{0.9}In_{0.1}Sb$ will be grown by Vertical Bridgman method with and without a magnetic field. The results will be compared with those obtained by further numerical simulation.

Acknowledgments

This research is supported by the European Community, through the INCO Strategic Action on Training and Excellence Program, contract number ICA1-CT-2002-70011 and through the TVP Cell Research and Training Network n’ HPRN-CT-2001-00199.

References


Authors

Prof. Duffar, Thierry  Dr.-Phys. Stelian, Carmen  Ph.D. student Mitric, Alina
EPM-MADYLAM/ENSHMG  West University of Timisoara  EPM-MADYLAM/ENSHMG
BP95  Bd.V.Parvan, No.4  BP95
38402 St Martin d'Hères, France  1900 Timisoara, Romania  38402 St Martin d'Hères, France
E-mail: thierry.duffar@inpg.fr  E-mail: carmen@physics.uvt.ro  Email: mitric@hmg.inpg.fr