Estimation of Diffusion Coefficient by Using Numerical Simulation of the Traveling Liquidus-zone Method

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Abstract

In order to estimate a diffusion coefficient, an InAs mole fraction profile, which is obtained by numerical simulation, is compared with a profile, which is experimentally obtained. The simulation result agrees well with the experimentally obtained profile. The diffusion coefficient is estimated to be about $1.1\times10^{-4}cm^2/s$ by comparing the numerically obtained growth length with the experimentally obtained one.

Introduction

The traveling liquidus-zone (TLZ) method[1, 2] can easily grow a homogeneous crystal of a ternary compound semiconductor such as InGaAs crystals. To grow a homogeneous crystal by the TLZ method, it is required to translate a sample by an appropriate translation rate. The translation rate is determined to be the same rate as a growth rate. The growth rate is estimated by using the one-dimensional TLZ model[3]. By using the TLZ method, single crystals of 2 mm in diameter and of more than 20 mm in length have been successfully obtained with good reproducibility on the ground[1, 2]. However, the one-dimensional TLZ model requires a diffusion coefficient. Therefore, to measure the diffusion coefficient, a microgravity experiment[4] was carried out by using a sounding rocket. From the experimental results, the coefficient and its temperature dependency was successfully obtained. However, it is often difficult to carry out the microgravity experiment to measure the diffusion coefficient. Therefore the diffusion coefficient is estimated by comparing a result from numerical simulation with an experimental result.

Two-dimensional Numerical Simulation

In this study, a numerically obtained InAs mole fraction profile is compared with an experimentally obtained profile. In order to obtain the profile, a two-dimensional numerical simulation is carried out. In the simulation, the energy transport equation, the mass transport equation, the energy balance equation and the mass balance equation are simultaneously solved. We apply this simulation to a sample with 2 mm in diameter. In the 2 mm sample, it has been already clarified that the convection effects on mass and thermal transports are negligibly small. Therefore, 2 mm sample is suitable for both the experimental estimation of the diffusion coefficient and the calculation time reduction. Thus we neglect the vorticity transport equation, the stream function equation and the terms related to the convection from the governing equations to shorten the calculation time. We use the boundary fitted coordinate

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(BFC) method[5-12], which is a kind of finite difference method, in order to solve these equations. The BFC method solves the governing equations that are transformed from the physical space to the computational space. The transformed governing equations are described as Eqs. (1) — (5).

$$\rho C_p \left\{ \frac{\partial T}{\partial t} - \frac{1}{J}(z_\eta T_\xi - z_\xi T_\eta)\frac{\partial r}{\partial t} - \frac{1}{J}(-r_\eta T_\xi + r_\xi T_\eta)\frac{\partial z}{\partial t} \right\},$$
$$= k\frac{1}{J^2}(\alpha T_{\xi\xi} - 2\beta T_{\xi\eta} + \gamma T_{\eta\eta}) + k\frac{1}{r}\frac{1}{J}(z_\eta T_\xi - z_\xi T_\eta) \tag{1}$$

$$\frac{\partial C_L}{\partial t} - \frac{1}{J}(z_\eta C_{L\xi} - z_\xi C_{L\eta})\frac{\partial r}{\partial t} - \frac{1}{J}(-r_\eta C_{L\xi} + r_\xi C_{L\eta})\frac{\partial z}{\partial t},$$
$$= D_L\frac{1}{J^2}(\alpha C_{L\xi\xi} - 2\beta C_{L\xi\eta} + \gamma C_{L\eta\eta}) + D_L\frac{1}{r}\frac{1}{J}(z_\eta C_{L\xi} - z_\xi C_{L\eta}) \tag{2}$$

$$\frac{\partial C_S}{\partial t} - \frac{1}{J}(z_\eta C_{S\xi} - z_\xi C_{S\eta})\frac{\partial r}{\partial t} - \frac{1}{J}(-r_\eta C_{S\xi} + r_\xi C_{S\eta})\frac{\partial z}{\partial t},$$
$$= D_S\frac{1}{J^2}(\alpha C_{S\xi\xi} - 2\beta C_{S\xi\eta} + \gamma C_{S\eta\eta}) + D_S\frac{1}{r}\frac{1}{J}(z_\eta C_{S\xi} - z_\xi C_{S\eta}) \tag{3}$$

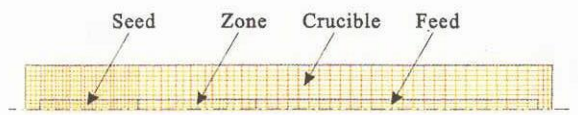
$$L_{SL}\rho\frac{\partial f}{\partial t}$$
$$= -k_L\frac{1}{r_\xi}\frac{1}{J_L}(-\beta T_\xi + \gamma T_\eta)_L ,$$
$$+ k_S\frac{1}{r_\xi}\frac{1}{J_S}(-\beta T_\xi + \gamma T_\eta)_S \tag{4}$$

$$(C_L - C_S)\frac{\partial f}{\partial t} ,$$
$$= -D_L\frac{1}{r_\xi}\frac{1}{J_L}(-\beta C_{L\xi} + \gamma C_{L\eta})_L \tag{5}$$

where, $\alpha = r_\eta^2 + z_\eta^2$, $\beta = r_\xi r_\eta + z_\xi z_\eta$, $\gamma = r_\xi^2 + z_\xi^2$, $J = r_\xi z_\eta - r_\eta z_\xi$, ξ and η are the computational coordinates corresponding to r and z in the physical space, ψ the stream function, ω vorticity, T temperature, ρ density, C_p specific heat, κ thermal conductivity, ν kinetic viscosity, L_{SL} latent heat, B thermal volume expansion coefficient, G the buoyancy coefficient by the specific gravity difference, g gravity, t time, C the concentration, D the diffusion coefficient. Subscripts of L and S indicate the liquid side and the solid side, respectively.

Results and Discussion

The generated grid and the initial configuration in this simulation are shown in Fig. 1 (a). The generated grid at the end of the growth is also shown in Fig. 1 (b). Although interface shapes are initially given as a flat shape, the shapes vary as the time evolves. This shape variation can be understood by comparing the initial grid with the final grid. As the temperature boundary condition, the temperature profile, which is shown in Fig. 2, is given on the crucible surface. The temperature profile moves towards the right side in Fig. 2 as the time evolves by the rate of 0.216 mm/hr, which is the experimentally optimized sample translation rate to grow a homogeneous $In_{0.3}Ga_{0.7}As$ crystal. Under these conditions, the numerical results are obtained. The typical result in the case of the diffusion coefficient of $1.2\times10^{-4}cm^2/s$ is shown in Fig. 3. In this figure, the red line and the blue one represent the InAs mole fraction measured by the electron probe micro-analyzer (EPMA) and the numerical result. As shown in Fig. 3, although the numerically obtained profile agrees well with the experimentally obtained one, the growth length obtained by the simulation is slightly longer then the experimental one. Therefore, another simulation in the case of the diffusion coefficient of $1.1\times10^{-4}cm^2/s$ is carried out. The comparison between the simulation and the experiment is shown in Fig. 4. This figure also shows that the InAs mole fraction profile and the growth length obtained from the simulation agree well with those from the experiment. Therefore, the diffusion coefficient of $In_{0.83}Ga_{0.17}As$ melt, which is the equilibrium composition to the solidus composition of $In_{0.3}Ga_{0.7}As$, is estimated to be $1.1\times10^{-4}cm^2/s$. In this simulation, the temperature at the growth interface is about 1293 K.



(a) Initial configuration

(b) Final configuration

Fig. 1 Generated grids and configurations

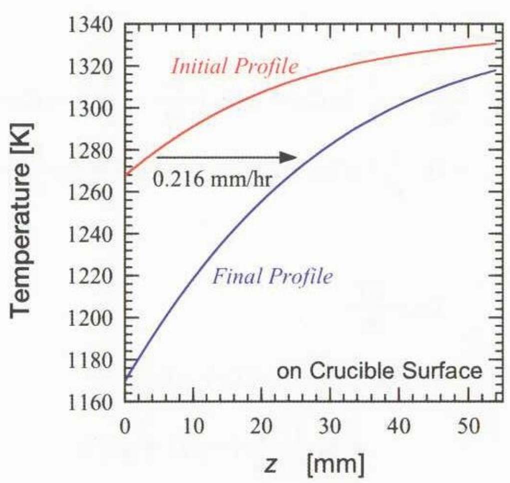


Fig. 2 Temperature profile

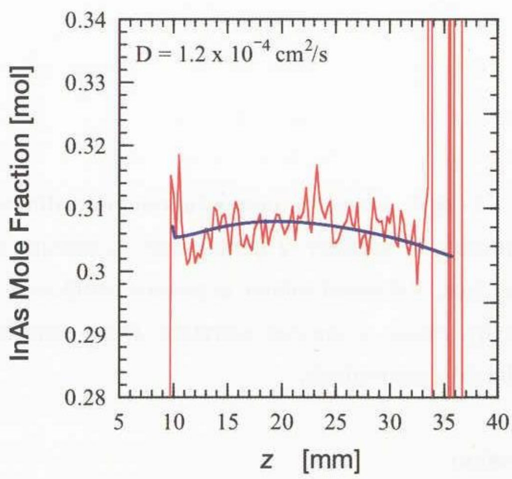


Fig. 3 Comparison between numerical and experimental results ($D = 1.2\times10^{-4}cm^2/s$)

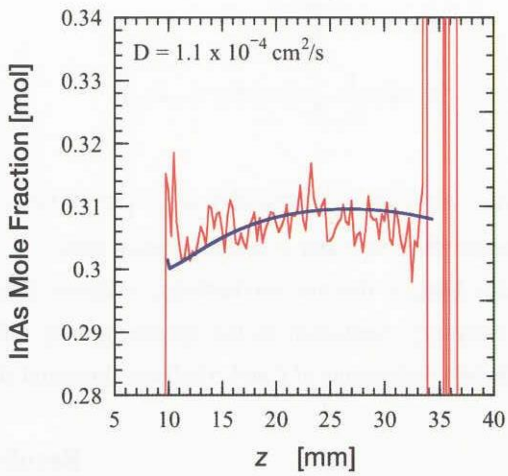


Fig. 4 Comparison between numerical and experimental results ($D = 1.1\times10^{-4}cm^2/s$)

In order to investigate the reliability of the estimation of the diffusion coefficient, the experimental results of the diffusion coefficient measurement by using the sounding rocket[4] is referred. The experimental results are shown in

Fig. 5. The natural logarithms of the measured coefficients are plotted in this figure. In Fig. 5, the minimum coefficient at each temperature is represented by a red closed circle. The middle one is represented by a blue closed circle. One scattered data is represented by a green closed square. The lower black line passes through the red circles and the upper one is the best-fitted linear line by using the blue circles. At 1293 K, the lower black line indicates that the diffusion coefficient is about $1.1\times10^{-4}cm^2/s$ and the upper black one indicates that the diffusion coefficient is about $1.4\times10^{-4}cm^2/s$. Therefore, the true diffusion coefficient may exist between $1.1\times10^{-4}cm^2/s$ and $1.4\times10^{-4}cm^2/s$. This is consistent with the estimation from the simulation. It is summarized that the estimation of the diffusion coefficient by the simulation is reliable enough.

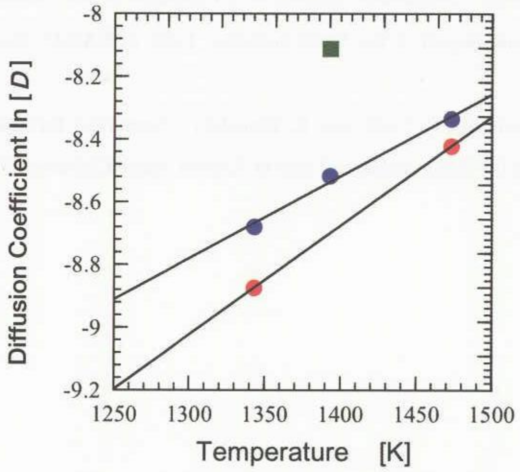


Fig. 5 Experimental result of diffusion coefficient measurement by using sounding rocket [4)]

Conclusions

In order to estimate a diffusion coefficient, numerical simulation is carried out. The InAs mole fraction profile obtained from the simulation is compared with that from the experiment. It is found that the numerically obtained profile agrees well with the experimentally obtained profile. In the case of the diffusion coefficient of $1.1\times10^{-4}cm^2/s$, the growth length also agrees well with the experimental result. Therefore, the diffusion coefficient of $In_{0.83}Ga_{0.17}As$ melt is estimated to be $1.1\times10^{-4}cm^2/s$ at 1293 K. To investigate the reliability of this estimation, the measurement results of the diffusion coefficient by using a sounding rocket is referred. From the experimental results, the diffusion coefficient is $1.1\times10^{-4}cm^2/s$ to $1.4\times10^{-4}cm^2/s$. This is consistent with the estimation by the simulation.

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