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Morphological Stability of Growth Interface from Solution in Semiconductor

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In this research, the morphological changes during GaP solution growth of the S/L interface were observed under static magnetic field using near-infrared (NIR) microscope with an interferometer. We discussed the damping effect of convection by means of analyzing fringe pattern during dissolution and growth.

Key words: In-situ observation, static magnetic field, liquid phase epitaxy, growth kinetics, morphological stability

1. INTRODUCTION

Flatness of crystal surface and interface is very important in order to produce quantum electronics devices and multilayer optical parts. The liquid phase epitaxy (LPE) growth method is one of the methods, [1] which produce high quality semiconductor crystals. Applications of extremely flat surfaces with low atomic step density are also important in surface physics. However, the problem of LPE is the inhomogeneous distribution of dopant caused by morphological instability of solid/liquid (S/L) interface during the crystal growth. Under the terrestrial gravity, it is very difficult to experimentally investigate the morphological stability due to the existence of convection. Therefore, it is necessary to control the heat and mass transports in fluid to be governed by diffusion.

In the present study, the magnetohydrodynamics effect by a static magnetic field is used to control the convection in semiconductor solution. The morphological changes of the S/L interface were observed in realtime using a near-infrared microscopic interferometer under the magnetic field, and the relationship between the growth condition and the morphological stability was evaluated.

2. EXPERIMENTAL

2.1 Crystal growth experimental

A schematic drawing of the experimental setup is shown in Figure 1. A GaP (111) substrate, a graphite crucible, 99.9999% Ga and a chromel-alumel thermocouple were put into a quartz glass ampoule. The ampoule was evacuated up to 10^{-3} Torr, and then sealed with a mechanical valve. The ampoule was set in the image furnace, and the temperature of the crucible was controlled by a PID temperature control system with the thermocouple. *In-situ* observation of back-melting and growth experiments by the linear heating and linear cooling were performed under a static magnetic field by using a superconducting magnet. The bright field image and interference fringe images were simultaneously obtained were recorded to a VTR system. Figure 2 shows the examples of bright field and interference fringe images under 5 T.

2.2 *In-situ* observation

The present experimental conditions are shown in Table 1. A semiconductor crystal has a transmittance for infrared ray whose wavelength is longer than the threshold wavelength. In this study, a near-infrared microscopic interferometer was designed based on the principle mentioned above. The reflected ray at the S/L interface was observed as a bright field image. The ray reflected from both the bottom of the substrate and the interface interferes and produce fringe image according to the thickness distribution of the substrate. The interference fringes correspond the contour line of the S/L interface with the height difference of 120 nm. [2]

3. RESULTS AND DISCUSSION

3.1 Stability of interface

Stability of the interface is calculated by a function of $f(\lambda)$ as

$$f(\lambda)=\frac{1}{\lambda^2(2\pi D+\beta_0\lambda)}\left[\left(\frac{C_S}{D}V_G-\frac{1}{m}G_T\right)\lambda^2-4\pi^2\Gamma C_{e0}\right], \quad (1)$$

where λ: wavelength of the macrostep, D: diffusion coefficient, β_0: kinetics coefficient of flat interface, C_S: concentration of the solute in crystals, V_G: growth rate, m: slope of the liquids line in the equilibrium phase diagram, G_T: temperature gradient at the interface and Γ: capillary constant. λ and V_G can be measured by the observation experiment. $f(\lambda)$ can be calculated with β_0 at the flat interface.

3.2 Back-melting and growth rate

The kinetics coefficient β_0 at the growth interface can be expressed by a mass conservation equations as follows,

$$C_S V_G(t)=D\left[\frac{\partial C(z,t)}{\partial z}\right]_Z=\beta_0\left[C_i(t)-C_e(t)\right], \quad (2)$$

where t : time, C: solute concentration of liquid, z : position variable, C_i: solute concentration and C_e: equilibrium concentration in liquid at the unperturbed

S/L interface.

At the one-dimensional diffusion-dominated model of LPE growth, the ratio of the growth rate V_G to the back-melting rate V_B is given by the following equation.

$$\frac{V_G(t)}{V_B(t)} = 1 - \frac{\sqrt{\pi}}{2\beta_0}\sqrt{\frac{D}{t}}\left\{1 - \exp\left(\beta_0^2\frac{t}{D}\right)\operatorname{erfc}\left(\beta_0\sqrt{\frac{t}{D}}\right)\right\}, \tag{3}$$

β_0 can be obtained as a result of curve-fitting of the measured value of $V_G(t)/V_B(t)$ by Eq.(3). As shown in Figure 3, V_G and V_B were measured from the interference fringe images obtained by the observation. $V_G(t)/V_B(t)$ is shown in Figure 4. The temperature dependence of β_0 obtained by Eq. 3 is shown in Figure 5. Wavelength of the macrostep calculated with the values of β_0 in Figure 5, $\lambda = 2\pi/\omega$, are shown in Fig. 6, where λ_0 and λ_{max} correspond to ω_0 and ω_{max}, respectively. The values of ω_0 and ω_{max}, at which $f(\omega_0) = 0$ and $f'(\omega_{max}) = 0$, characterize the behavior of the undulation: the growth is unstable for $\omega < \omega_0$ and the growth surface may be covered with the wave $\omega = \omega_{max}$. The measured λ satisfied the relationship that $\lambda > \lambda_0$ and $\lambda \sim \lambda_{max}$. The deviation of λ from λ_{max} may be attributed to the assumption where the macrostep has a sinusoidal shape.

4. CONCLUSION

The kinetics coefficient on the vicinal surface in LPE growth of GaP/GaP(111)B was obtained by an *in situ* observation technique under the strongly reduced convection condition. The calculated value of macrostep wavelength agreed well with the measured one, and therefore the validity of the present model is considered to be suitable for explaining the morphological stability of the S/L interface in LPE growth of semiconductor crystals.

REFERENCES

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[2] D. Yin and Y. Inatomi, *Cryst. Res. and Technol.* **35** (2000) 221.

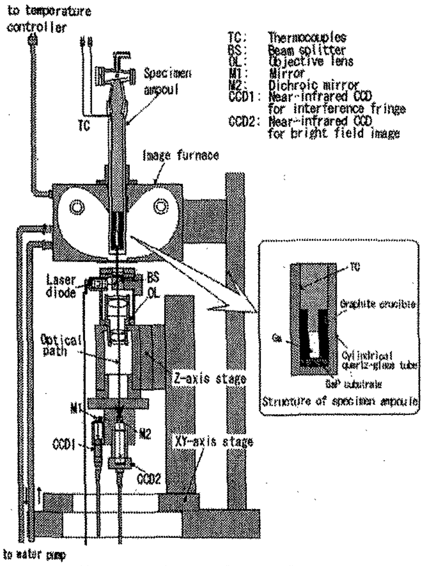

Figure 1. Experimental setup

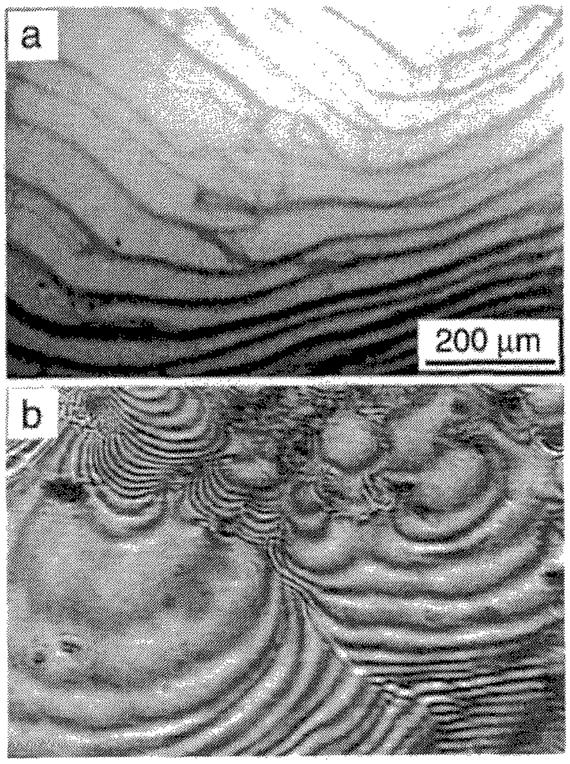

Figure 2. Typical observed images of S/L interface during GaP/Ga(111)B LPE growth by NIR-MI: (a) a bright field image of macrosteps, (b) an interference fringe image of small facet regions.

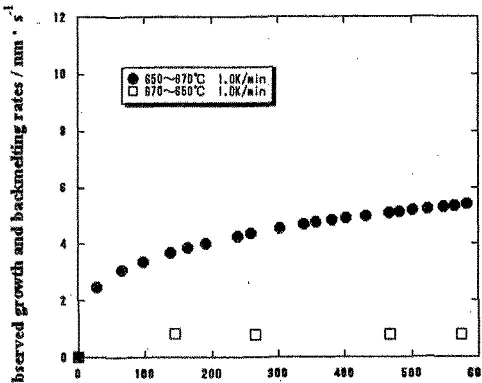

Figure 3. Growth and Back-melting rate as functions of time under 5 T.

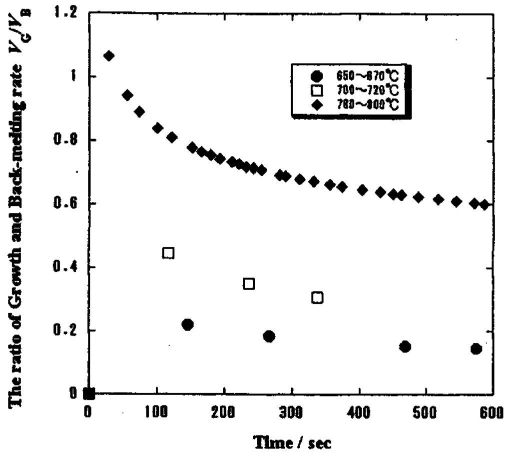

Figure 4. The ratio of Growth rate and Back-melting rate.

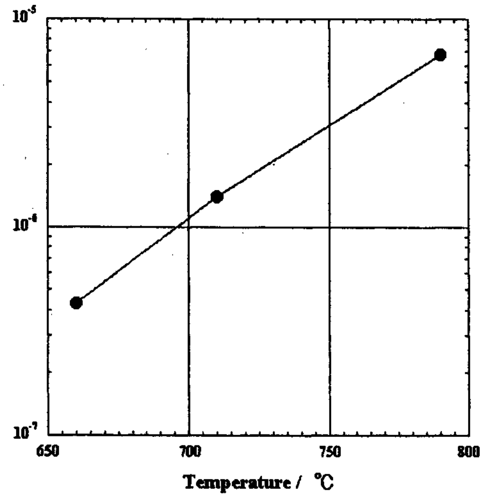

Figure 5. The temperature dependence of kinetics coefficient.

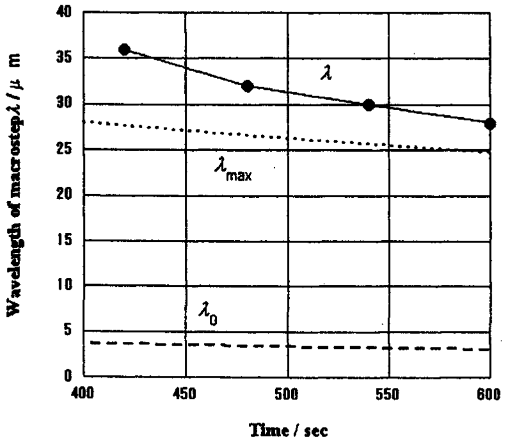

Figure 6. Wavelength of the macrostep λ as function of time.

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