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In-situ observation of semiconductor crystal growth from solution in a high static magnetic field

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In this research, the morphological changes during GaP solution growth of the S/L interface were observed under strong 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, natural convection, diffusion-controlled model, static magnetic field

1. Introduction

For the high quality of semiconductor crystal, it is necessary to make clear the phenomena of morphological change of the solid/liquid (S/L) interface, *e.g.*, a mechanism of shaping macrosteps. The macrostep causes inhomogeneous dopant distribution, and its segregation trace of the dopants is one of the barriers to produce a high quality crystal for optoelectronics devices.

Mass and heat transport phenomena in solution during growth is governed by thermal and solutal convections. These convections were thought to cause the homogeneities of temperature and solute concentration distributions near the S/L interface. Therefore, it is difficult to understand the phenomena during process under the terrestrial gravity because of nature convection in the liquid phase.

The control of natural convection is a one of the promising method to control component and defect distributions for materials processing. In order to suppress the convection in solution, a static magnetic field is used as a pseudo-microgravity environment for conductive flows [1-3]. In the present research, the morphological changes of the S/L interface during GaP solution growth were observed under strong static magnetic field using a near-infrared (NIR) microscopic interferometer.

2. Experimental procedure

We developed the experiment setup to observe the S/L interface under high static magnetic field. By utilizing the transparent properties of semiconductors for infrared radiation, we succeeded in directly observing the morphological changes of the solid-liquid interface during solution growth in GaAs/GaAs [4], CdTe/CdTe [4], GaP/GaP [4] and InP/InP [5] using the infrared microscopic interferometer. This optical system was adjusted to focus on the solid-liquid interface through the substrate placed inside the tube.

The both of the fringe pattern and the bright field image could be observe simultaneously. The displacement of the S/L interface was measured by counting the amount of the interference fringe displacement on the facet region using a NIR microscopic interferometer. A 5 mW laser diode with wavelength of λ = 780 nm was used to a light source for the interferometer [3]. An interference fringe corresponds to 120 nm in height difference of S/L interface. The image beam reflected at the S/L interface was received by a CCD camera, and digitally calculated and recorded to a computer through an image processor. The electrical function of the camera is influenced less by a strong magnetic field than by that of an infrared image pickup tube.

The substrate used for the experiment was S-doped GaP crystals, 300 μm in thickness and ϕ 7.8 mm, with orientations of (111)B $\pm$ 0.1°. Both sides of substrate were mechanically and chemically polished. Solution growth experiments were performed in a quartz glass ampoule consisting a graphite crucible, a GaP substrate and a Ga solvent. The ampoules were evacuated up to 10^{-5} torr, and set at the center of a ellipsoidal image furnace in a superconducting magnet whose maximum magnetic induction, *B*, was 6 T. The schematic drawing and the photo of the setup are shown in Fig. 1.

The lamp power of the furnace was controlled by a DC power supply. The dissolution and growth procedures were carried out by linear heating and cooling methods with the rates of 0.5 K/min, respectively. The temperature range for the procedures was from 923 to 933 K. To sufficiently saturate the solute distribution in the liquid before growth was started, *B* was held at 0 T for 2 hours to stir the liquid with the help of the convective flow currents, and then at the target value of *B* for an additional 2 hours. We succeeded in observation of a bright field image and a fringe pattern in 6 T as shown in Figs. 2 and 3.

3. Results and Discussion

The influence of the buoyancy force on natural convection was studied by Camel and Favier using a scaling analysis.[6] In this study, the same analysis was applied to the Navier-Stokes equation taking account of thermal convection and Lorenz force in order to estimate the fluid behavior in magnetic field as follows:

$$\mathrm{Re}^2 + (1+\mathrm{Ha}^2)\mathrm{Re} \approx \left| m(\mathrm{Gr_T}+\mathrm{Gr_S}) \right|, \qquad (1)$$

where Re: Reynolds number = $u_r r/\nu$, Ha: Hartmann number = $Br(\sigma/\rho\nu)^{1/2}$, $\mathrm{Gr_T}$: thermal Grashof number = $\beta G_T g_0 r^3/\nu^2$, $\mathrm{Gr_T}$: solutal Grashof number = $\alpha G_C g_0 r^3/\nu^2$, u_r: characteristic velocity in the solution, ν: kinematic viscosity of the solution, r: radius of the solution, σ: electrical conductivity of the solution, ρ: denisty of the solution, G_T and G_C: axial temperature and concentration gradients in the vicinity of the solid/liquid interface, and m: the gradient of the S/L interface.

In Eq. (1), the solutal convection can be neglected, because the substrate has the concentration of phosphorus less than 1 at. %. A hypothetical low-gravity level, g, in which Re was the same value as that in static magnetic induction B, was estimated based on the Eq. (1). The relation of g and B is shown in Fig. 4. When we assumed the gravity level g was 10^{-4} g_0 (g_0 = 9.8 m/s^2) as in the space shuttle, an induction of 2 T simulated the same flow behavior, and the condition at B = 6 T is enough to damp the convection to be a diffusive mass transport state.

During growth, hexagonal-shaped hillocks generated at the dislocations in the substrate appeared at B = 0 T. On the other hand, the large facet region with few hillocks was observed at 6 T as shown in Fig. 2. It was thought that the morphology of the growth interface at 6 T was more stable than that at 0 T. The curves of the dissolution and growth thicknesses at 6 T, shown in Fig. 5, were close to the calculated values, H(t), based diffusion-controlled model:

$$H(t) = \frac{Lp}{C_S}\left[t + 2\sum_{n=1}^{\infty} \frac{L}{a_n^4 D} \exp\left(-\frac{a_n^2 D}{L} t \right) \right], \qquad (2)$$

where L: length of the solution, C_S: concentration of the solute in the solid, p: (dCe/dt) as a constant, Ce: equilibrium concentration in the solution at the S/L/ interface, and $a_n = (2n-1)\pi/2$, respectively. [7)]

4. Conclusion

A nearly diffusion-controlled condition in the liquid was produced using an axial magnetic induction of 6 T. The morphology of the S/L interface was stabilized by damping of the convective flow due to the strong static magnetic field.

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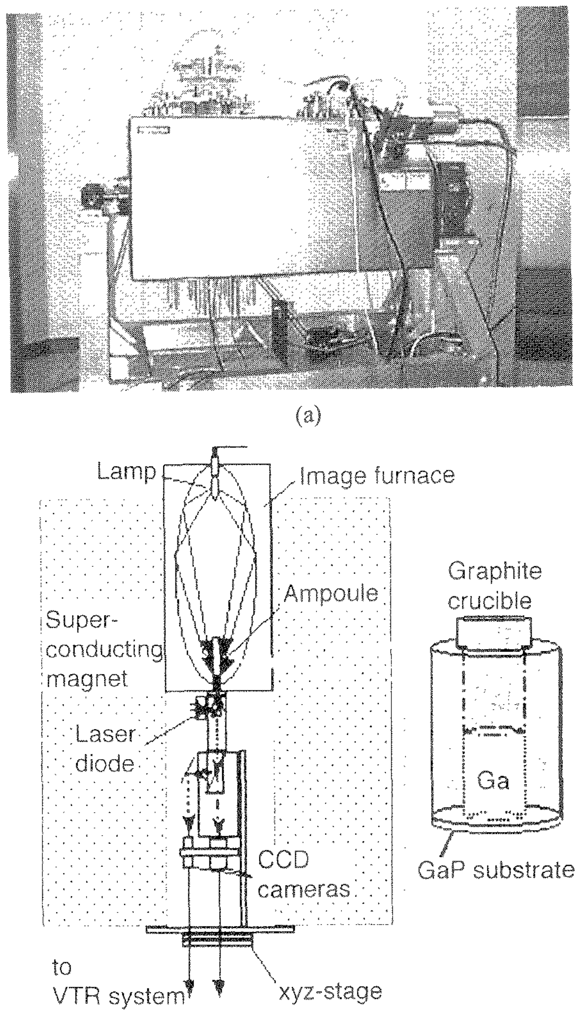

Fig. 1. The experimental setup: (a) a photo, and (b) a schematic drawing.

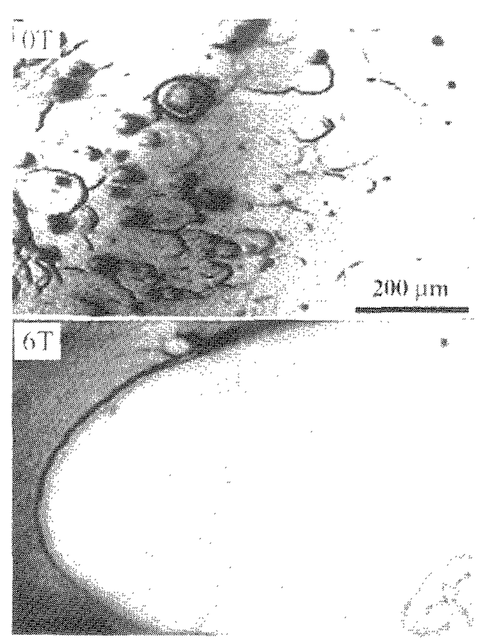

Fig. 2. The bright field image of the growing S/L interface on the facet region.

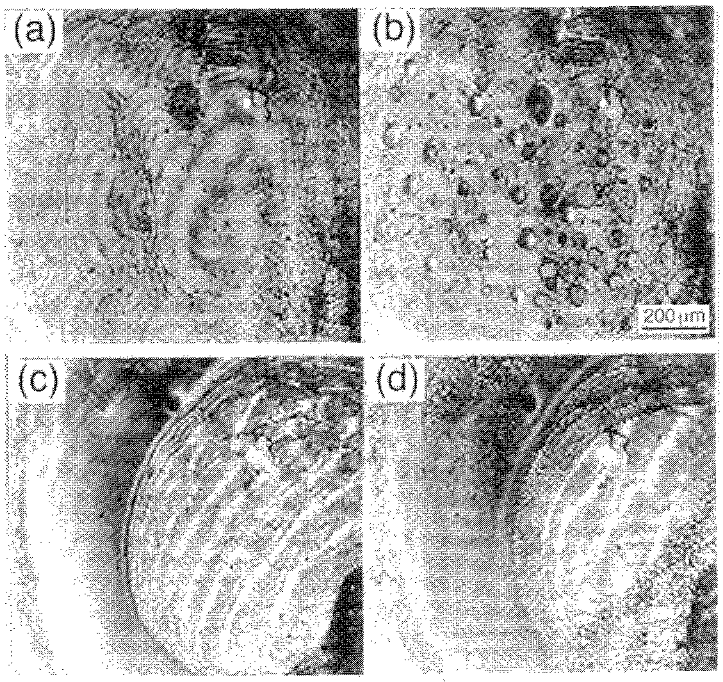

Fig. 3. The fringe pattern of the S/L interface on the facet region: (a) 0 T, before growth (t = 0 min), (b) 0 T, during growth (t = 20 min), c) 6 T, before growth (t = 0 min), (d) 6 T, during growth (t = 20 min).

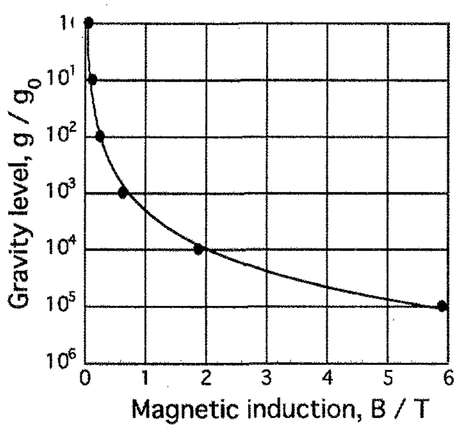

Fig. 4. The relation between hypothetical gravity levels with magnetic induction.

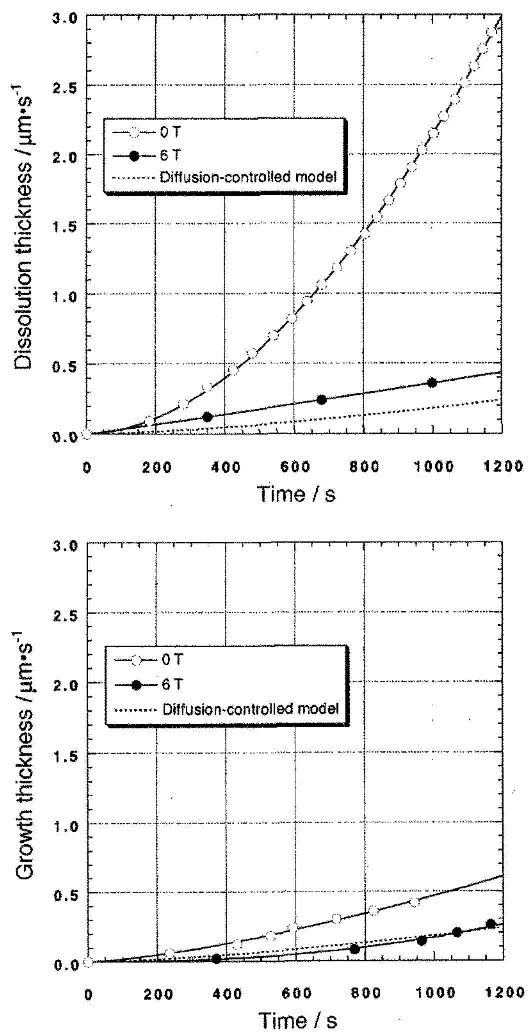

Fig. 5. The dissolution and growth thicknesses as functions of time.

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