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Lateral Growth Mechanisms of GaAs MOCVD Layers

Hiromitsu Asai

NTT Electrical Communications Laboratories,

3-9-11 Midori-cho, Musashino-shi, Tokyo 180, Japan

For metalorganic chemical vapor deposition, anisotropy in lateral growth is observed on (001) GaAs having round mesas. The [110] growth rate is higher than the [110] under high As pressure. The [110] growth rate decreases with decreasing As pressure while the [110] remains constant. As growth temperature increases, both the [110] and [110] growth rates decrease. These results are well explained by a "bond number model" based on atomic arrangements and the number of dangling bonds at [110] and [110] step sites.

I. INTRODUCTION

Epitaxial growth on preferentially etched and/or masked substrates is necessary to fabricate buried-type-structure devices, such as lateral confinement lasers1) and permeable base transistors.^{2,3)} For this purpose, metalorganic chemical vapor deposition (MOCVD) is one of the most suitable techniques because of its high selectivity and high tolerance to contamination at a re-growth interface.⁴⁻⁶⁾ In order to successfully fabricate device structures requiring lateral control of composition, doping, and geometrical shape, lateral growth processes must be understood. In addition to this technical view point, the investigation on the lateral growth processes will allow understanding of surface kinetics in basic growth mechanisms. Nishizawa et al.⁷⁾ have observed a high lateral growth rate for chloride vapor phase epitaxy (chloride-VPE) of Si on (111) substrates having round mesas, and have examined its growth mechanisms. For GaAs MOCVD, however, lateral growth processes have not yet been studied. The purpose of this work is to clarify the lateral growth mechanisms in GaAs MOCVD layers on (001) substrates, with a method similar to that reported by Nishizawa et al.⁷⁾

In this paper, the anisotropy in lateral growth on (001) surfaces and its dependence on growth conditions are described for MOCVD growth for the first time. Growth temperature and As partial pressure was found to have strong influence on the lateral growth rates but not on the vertical ones. On the basis of these results, we propose a "bond number model" to clarify the lateral growth processes.

II. EXPERIMENTAL PROCEDURE

The MOCVD system used in this work was composed of a horizontal quartz reactor, an rf induction heater, and a high-purity carbon susceptor. The upper surface of the carbon susceptor was tilted about 7^{0} from the reactor tube. Trimethylgallium (TMG) and arsine (AsH₃) were used as source gases to grow GaAs. A purified H₂ carrier gas having 4 l/min flow rate was introduced with the TMG and AsH₃ into the reactor. Growth temperature was monitored with a thermocouple inserted into the carbon susceptor. Growth was performed under atmospheric pressure.

The substrates were Si-doped GaAs $(001)\pm0.5^{\circ}$ wafers. After etching the wafer with a $3H_2SO_4$ -



Fig.1. Schematic picture of substrate.



Fig.2. Surface photographs (a) before and (b) after growth.

1H₂O₂-1H₂O solution, a 10-um diameter circular mesa having a 1200-A step height, as shown in Fig. 1, was made on the wafer by a conventional photolithographic technique and wet etching. The lateral growth rates were determined by measuring the spread of the circular mesa edge after 1 hr growth, and vertical growth rates were measured by stain-etching.

III. RESULTS

Typical surface photographs before and after 1 hr growth on circular mesas are shown in Fig. 2. It is clear from this figure that lateral growth has anisotropy between the [110] and [I10] directions. The [110] growth rate was the highest in all directions on the (001) surface, while the [I10] growth rate was the lowest. Note that lateral spreads from the original mesa edges (10 um and 5 um for [110] and [I10], respectively) were very large in comparison with the 1.8-um layer thickness.

The dependence of [110] and [I10] growth rates on the growth temperature is presented in Fig. 3. The growth temperature was varied from 550°C to 800°C, while the other growth parameters were held constant. Although the vertical growth rate was independent of the growth temperature, the lateral growth rate exhibited strong temperature dependence. The [110] growth rate remained 9 um/hr up to 650°C and subsequently decreased as the temperature increased. On the other hand, the [I10] growth rate was 5 um/hr up to 700°C and then it decreased. It should be noted that both the [110] and [T10] growth rates decrease with temperature in contrast to Si chloride-VPE. 7) This indicates that the lateral growth processes in MOCVD are not limited by surface atom diffusion.



Fig.3. Lateral and vertical growth rates as a function of growth temperature.

The arsenic partial pressure ([AsH₃]) also has influence on the lateral growth. The [AsH₃] dependence of [110] and [I10] growth rates under constant [TMG] is represented in Fig. 4. The [I10] growth rate was almost constant in the [AsH₃] range of $1.1x10^{-4}$ to $3.7x10^{-3}$ atm, corresponding to [AsH₃]/[TMG]= 1.7-58. On the other hand, the [110] growth rate increased with increasing an [AsH₃]. However, the increment tends to be saturated above [AsH₃] of $1.8x10^{-3}$ atm. It is noteworthy that crossing between the [110] and [I10] growth rates occurs around [AsH₃] of $4.0x10^{-4}$ atm.



Fig.4. Lateral and vertical growth rates as a function of [AsH₃].

Figure 5 shows the ratios of [110] and [I10] growth rates to vertical one as a function of [TMG]. Both the ratios is found to be independent of [TMG]. Since the vertical growth rate is well known to increase linearly with an increase in [TMG], the [110] and [I10] growth rates also have linear dependence.

IV. DISCUSSION

The marked results obtained in this work are summarized as follows; (1) [110] growth rate is higher than for [I10], (2) [110] and [I10] growth rates decrease with growth temperature, (3) [110] growth rate decreases as [AsH₃] decreases while [I10] growth rate remains almost constant.

For Si chloride-VPE, a high lateral growth rate has been reported to follow that the Si sticking probability into atomic steps or kinks is higher than that on flat surfaces.⁷⁾ However, anisotropy between [110] and [T10] growth rates obtained for GaAs MOCVD can not be explained by this simple atomic step model. This is because the model takes no account of the differences in Ga and As atomic arrangements and the number of dangling bonds at the [110] and [T10] steps.

We now propose a "bond number model" to explain the lateral growth processes, on the basis of the foregoing experimental results. A basic assumption is that the lateral growth rate is proportional to the number of bonds for binding a Ga atom



Fig.5. Ratio of lateral to vertical growth rates as a function of [TMG].



Fig.6. Schematic pictures of (a) [110] and (b) [110] steps.

to the growing step edge. This model can explain the anisotropy in lateral growth rates and its dependence on the growth conditions.

Under high As pressure, a growing surface is considered to be completely covered with As species. Cross sections of [110] and [I10] steps under this condition are schematically drawn in Fig. 6. The Ga atom at the [110] step (Gat1101) is bound with three bonds; one to the step side and two to the step bottom, while the Gattion is bound with two bonds; both to the step bottom. Assuming that the incorporation probabilities of Ga species into the steps is proportional to the number of bonds for binding to the steps, Ga atoms, which are migrating on the growing surface, are easier to be incorporated into the [110] steps than into the [I10]. As a consequence, the [110] growth rate is essentially higher than the [I10] one under high As pressure (see Fig. 3).

Under decreasing [AsH₃], the As \dagger_{110} must be easily desorbed in comparison with the As \dagger_{110} . This is because the As \dagger_{110} is bound to the growing step with only one bond, as shown in Fig. 6(a). The number of bonds for Ga \dagger_{110} changes from three to two when the As \dagger_{110} is desorbed, although that for Ga \dagger_{110} remains two. As a consequence, the [110] growth rate reduces with a decrease in [AsH₃] while the [I10] one is almost independent.

At higher temperature, the $As^{\dagger}_{I10]}$ also begins to be desorbed. Under this condition, both the $Ga^{\dagger}_{110]}$ and $Ga^{\dagger}_{I10]}$ might be stuck, not to the step sites but on the flat surface. Therefore, the [110] and [I10] growth rates decrease in the high temperature range (Fig. 3). In addition, since As[†]_{110]} desorption occurs at a relatively low temperature, the temperature at which the [110] growth rate begins to decrease is slightly lower compared with that for the [I10] growth rate.

Assuming that the As adsorption/desorption processes at the step sites obey the Langmuir adsorption formula and that migrating Ga species react with the adsorbed As,⁸⁾ the lateral growth rate, $R_{lateral}$, is given by the equation,

$$R_{lateral} = k \frac{B_{As} \exp(E_{As}/k_{B}T) P_{As}}{1 + B_{As} \exp(E_{As}/k_{B}T) P_{As}} P_{Ga}, \quad (1)$$

where k is a constant, B_{AS} is a pre-exponential factor, E_{AS} indicates adsorption energy for As species, and P_{AS} and P_{Ga} denotes the partial pressure for As and Ga species, respectively. Note that K includes the incorporation probability for Ga species into step sites.

It is clear from Eq. (1) that the lateral growth rate depends linearly on [TMG] similarly to the vertical growth rate (see Fig. 5). Adjusting the parameters of k, $B_{\mbox{AS}},$ and $E_{\mbox{AS}},$ fit curves for experimental data of the [110] and [I10] growth rates can be obtained. They are shown as solid lines in Figs. 3 and 4. In this case, EAs's at the [110] and [I10] steps were estimated at 2.32 eV and 4.64 eV, respectively. Considering that the Astiloj and Astiloj are bound with one and two bonds, respectively, it is reasonable that E_{As} at the [I10] is twice as large as the one at the [110]. In addition, these values are also considered to be valid, compared with the cohesive energy for GaAs (6.33 eV).9) In this way, the "bond number model" proposed here can explain well our experimental results. This model will be successfully applied to the lateral growth mechanisms for other zincblende-type-semiconductors.

V. CONCLUSION

Anisotropic lateral growth was observed in GaAs MOCVD layers on (001) substrates having circular mesas. The [110] growth rate was higher than the [I10] one under high As pressure. The lateral growth rates depended on the growth temperature and the [AsH₃], although the vertical growth rate was independent. The [110] growth rate decreased with an increase in growth temperature or a decrease in [AsH₃], with stronger dependency than that for the [I10] growth rate. These results were well explained by the "bond number model" proposed here.

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