Novel Step Height Reduction Phenomenon during Alkyl-Desorption Limited Atomic Layer Epitaxial Growth on Vicinal Substrate

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The behavior of the step height during an atomic layer epitaxial (ALE) growth of GaAs on vicinal substrates is studied by atomic force microscope (AFM). A new alkyl-desorption limited (ADL) ALE mode recently found by our group is used, where desorption of alkyl, rather than saturated adsorption of alkyl, realizes one ML/cycle growth. It is found that step heights reduce during the ADL-ALE, and this new phenomenon is explained by the ADL-ALE mechanism.

1. Introduction

If artificial control of step-heights is possible, fabrication of defect-free quantum structures in a self-organizing fashion becomes possible by preferential growth on mono- or multi-atomic steps. Although step bunching and formation of multi-atomic steps are commonly observed in the ordinary MOVPE growth, no report has been made on the behavior of steps during atomic layer epitaxial (ALE) growth which is an attractive technique to control the growth with precision of one monolayer (ML).

The purpose of this paper is to study, by atomic force microscope (AFM), the behavior of the step height during the alkyl-desorption limited (ADL) ALE growth of GaAs using TEGa and AsH₃ on vicinal substrates.

ADL-ALE is a new ALE mode recently found by our group¹⁾ where desorption of alkyl, rather than saturated adsorption of alkyl, realizes one ML/cycle growth. Conventional modes of ALE are known to have only specific narrow temperature ranges $^{2),3)}$ for ALE which makes it difficult specific narrow temperature exploit to them for growth of heterostructures. As opposed to this, only the ADL-ALE mode has so far achieved ALE growth of GaAs and InAs at the same growth temperature.

It is shown here for the first time that step heights reduce during ADL-ALE growth contrary to the MOVPE growth. This new phenomenon, which is explained here by the ADL-ALE mechanism, may open up a possibility of artificial control of step heights by combining it with step height increase in the conventional MOVPE growth.

2. Alkyl-desorption limited ALE

Figure 1 shows schematically the behavior of the growth rate in the ADL-ALE mode. The growth rate does not saturate with TEGa supply duration, but shows an initial rapid reaction region (rapid region), followed by a subsequent slow reaction region (slow region). However, clear saturation of the growth rate to 1ML/cycle takes place against various pressures of TEGa supply if the pulse duration of TEGa is held appropriately fixed. A similar growth mode can be utilized for ALE growth of InAs using trimethylindium (TMIn) and AsH₃.



Fig.1 Schematic behavior of the growth rate in the ADL-ALE mode.

3. Experimental

The growth system used in the present work was an atmospheric pressure MOVPE system with a vertical reactor. The vertical reactor consisted of 50mm innerdiameter quartz tube with water-cooling jacket and carbon susceptor. The susceptor was heated by an RF induction coil. Substrates with a size of 10mm x 10mm were placed on the carbon susceptor with their surfaces inclined at about 30⁰ to the gas flow direction so as not to disturb the gas flow pattern. The source materials were TEGa, TMIn, trimethylaluminium (TMAl) and 10% AsH₃ in H₂. Hydrogen was used as the carrier gas and the total flow rate was kept to 6.0 standard litter per minute (SLM). Samples were grown on semi-insulating (001) GaAs substrates misoriented towards <110> and <110> direction with angle of misorientation 2.0°. In order to maintain well defined initial surface conditions, carefully prepared GaAs bulk wafers with specified misorientation and 2-3 atomic steps were used.

In order to investigate the step height change during the ADL-ALE growth, two types of samples which had different initial surfaces with different initial average step heights or average step periodicity were prepared. One type of the sample had a GaAs buffer layer of a thickness of 1000Å as the initial surface (Type 1 Samples), and type had an the other AlAs/GaAs superlattice (SL) structure of 100 periods on a GaAs buffer layer of a thickness of 1000Å (Type 2 Samples). Both of the GaAs buffer and superlattice were grown by the conventional MOVPE mode at 650°C.

The surface images of samples were obtained with an atomic force microscope (Digital Instruments Nanoscope II). A silicon nitride integrated tip was used and the measurements were done in the air. No special sample treatments were carried out before imaging.

4. Results and discussion

Figure 2 shows the observed average step periodicity vs. grown thickness (i) for repeated growth in the rapid region of ADL-ALE growth and (ii) for repeated growth in the slow region of ADL-ALE growth at a growth rate of 1ML/cycle. The initial step periodicity was 430 nm (Type 1 Sample) and the ADL-ALE growth of GaAs was done at the temperature of 310^oC. After the growth in the slow region, a marked reduction of step height is observed. On the other hand, no change of step height took place after the growth in the rapid region.

Figure 3 shows the result of a similar experiment in the slow region at the same growth temperature, but starting from a much smaller initial step periodicity of 85 nm (Type 2 Sample). Again, clear reduction of step height is observed. The step height seems to saturate to a final value of about 50nm.



Fig.2 Observed average step periodicity vs. grown thickness. Initial step periodicity was 430 nm.



Fig.3 Observed average step periodicity vs. grown thickness. Initial step periodicity was 85 nm.

In order to see the microstructure of the ADL-ALE grown layer near the steps, 2ML of InAs was grown by ADL-ALE at $305^{\circ}C$ on a GaAs vicinal surface, and capped by a top GaAs layer. Figure 4 shows the cross-sectional transmission electron microscopy (TEM) image of the sample. It is seen that the InAs thickness becomes obviously larger near steps, indicating accumulation of In atoms during ADL-ALE growth of InAs.



Fig.4 Cross-sectional TEM image of the ADL-ALE grown InAs.

The above results can be explained in terms of our recently proposed ADL-ALE mechanism shown in Fig.5(a) where adsorption of alkyl-Ga proceeds in the rapid region until it is limited by steric hindrance, followed by gradual growth limited by alkyl desorption in the slow region. Thus, no change of step height is expected for repeated growth in the rapid region. On the other hand, the likely situation near step in the slow region is shown in Fig.5(b). Since the alkyl desorption rate on the steps seems to be much higher than that on the terraces, excess Ga atoms accumulate at the step sites during TEGa supply. Then, in the subsequent AsH₃ supply cycle, excess Ga atoms diffuse onto the terrace region to form GaAs in a way similar to the recently reported behavior of Ga droplet in MEE growth⁴⁾. This obviously leads to reduction step heights. The occurrence of of saturation is consistent with such a model, since amount of Ga accumulation itself reduces with the reduction of step heights.







Fig.5 (b) Growth model near step.

Conclusion

The behavior of the step height during the ADL-ALE growth of GaAs using TEGa and AsH₃ on vicinal substrates is studied by AFM. Step heights reduce during the ADL-ALE, and this new phenomenon is explained by the ADL-ALE mechanism.

References

 S.Goto, J.Ishizaki, T.Fukui and H.Hasegawa, Int. Symp. GaAs and Rerated Compounds, Karuizawa, 1992, 139.
J.Nishizawa, T.Kurabayashi, H.Abe and N.Sakurai, J.Vac.Sci.Technol., <u>A5</u>(1987)1572
H.Ohno, S.Ohtsuka, H.Ishii, Y.Matsubara and H.Hasegawa, Jpn.J.Apll.Phys.,<u>54</u>(1989)2000
J.Osaka, N.Inoue,Y.Mada, K.Yamada and K.Wada: J. Crystal Growth, <u>99</u>(1990)120.